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Temporal Analysis of Radiocesium Concentration in Sewage Sludge after Fukushima Daichi Nuclear Power Plant Accident

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Abstract. Fukushima Dai-ichi Nuclear Power Plant (FDNPP) accident in April 2011 had released radioactive substances to the environment. Radiocesium transported from the urban area to the sewerage system by transfer wash-off mechanism. Eventually, radiocesium accumulated in the sludge produced by the wastewater treatment plant. This study was aimed to evaluate the possibility of using the time-series of Radiocesium concentrations in sewage sludge to quantify the wash off rate of the radionuclide on the surface of an urban area. This study used monitoring data from four WWTPs in Fukushima Prefecture and daily concentrations of Radiocesium in their sewer sludge over 8 years were collected. The results show time-dependent of Radiocesium concentration in sludge fitted with double exponential regression which means there are two rates: rapid rate (22 – 36 months) and slow rate (123 – 213 months). Values of the model parameters based on temporal analysis of radiocesium concentrations in sewage sludge are consistent with radiocesium wash off after the Chernobyl accident indicating that the decreasing rate of radiocesium concentrations in sewage sludge reflect the wash off rates on the surface of the corresponding urban area.

1. Introduction

Fukushima Daichi Nuclear Power Plant (FDNPP) accident had released a substantial amount of radiocesium into the environment. As a consequence, urban areas surrounding the power plant received the deposition of the radionuclide and inhabitants living in surrounding areas of FDNPP were evacuated to prevent health impact due to radiation exposure. In urban areas, the flux of radiocesium redistribution might be temporally stored in a drainage and sewer system before it is discharged to a receiving water body. The flows of surface run-off and municipal sewage in an urban area typically managed by a drainage and a sewer system or a combined system. This system is deployed to prevent the pollution of the receiving water body due to the contaminant contained in the both surface run-off and municipal sewage. The pollution control is carried out in a WWTP by separating the contaminant from the wastewater and producing sewer sludge. When the surface water and municipal sewage contains radiocesium, the redistribution process would involve the drainage and sewer system. Moreover, due to the separation process in WWTP, it separates some fraction of radionuclide and accumulates it in the sewer sludge [1]. According to monitoring activities conducted by the local prefectural government, about 50-11,000 Bq.kg^{-1}was found from the sludge from a WWTP in Fukushima City in two years after the accident [2].

Temporal analysis of radiocesium redistribution in urban areas is an important information after a major nuclear accident as it determines the rate of natural decontamination due to hydrological process. After the deposition of the radionuclide on the ground surface, some fraction of it would be rapidly removed and the remaining part would be migrated in a very long period. The first part is generally known as rapid wash off whereas the later part is called slow wash off. Rapid wash off occurs during days or months after the accident. The slow wash off due to the fixation of the radionuclide to soil particle would last in decades depending on the fixation capacity and the natural decay rate of the radionuclide [3].

Data Collection

| ^{137}Cs and ^{134}Cs deposition (D) | ^{137}Cs and ^{134}Cs concentration in sewage sludge (c) | WWTP operational data |

Temporal analysis

Model 1	$c(t) = D(Be^{-k_1 t})$
Model 2	$c(t) = D(Be^{-k_1 t} + Ce^{-k_2 t})$
Model 3	$c(t) = D(A + Be^{-k_1 t} + Ce^{-k_2 t})$

Model evaluation:
1. Determination coefficient (R^2)
2. Standard error of estimate (E)

Model parameter quantification

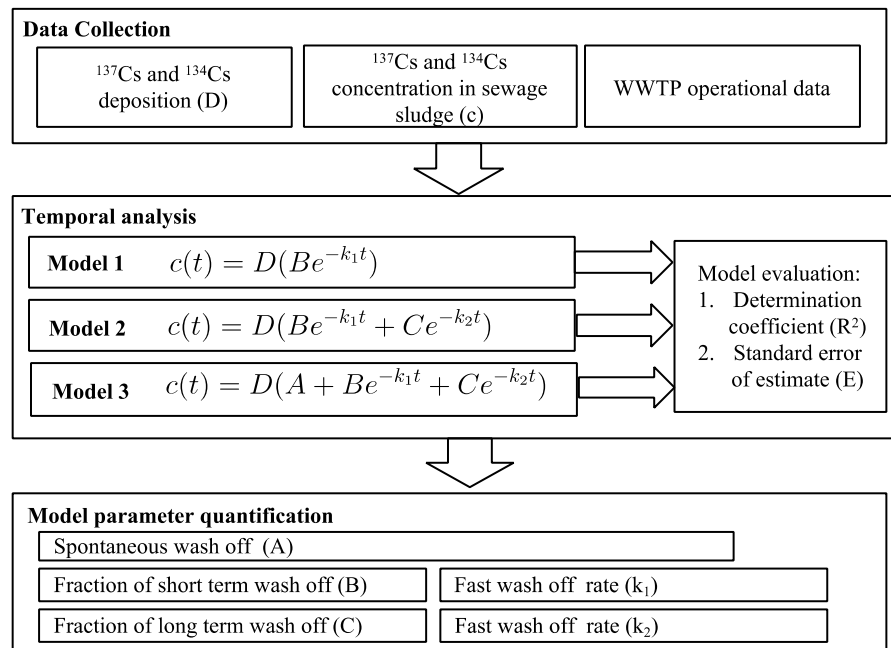| Spontaneous wash off (A) | |
| Fraction of short term wash off (B) | Fast wash off rate (k_1) |
| Fraction of long term wash off (C) | Fast wash off rate (k_2) |

Figure1. Framework of the study consisting of three steps. The data collection step is followed by temporal analysis to evaluate and choose the best fit model to the observed data of ^{137}Cs and ^{134}Cs concentration in sewage sludge. Finally, the parameter values of the chosen model were quantified.

Values of rapid and slow wash off rate of several radionuclides have been studied throughout various river catchments during three major radionuclide release events (Weapon Test era, Chernobyl accident and Fukushima accident [4–6]. The challenge of these studies was the difficulties of collecting radiocesium concentration data not only in spatial wise as the extensiveness of a major river catchments area but also in temporal wise as it requires frequent monitoring data in a long period. On the other hand, after the FDNPP accident, the local prefectural government of Fukushima has been releasing their daily monthly monitoring data of radiocesium concentrations in sewage sludge taken from WWTPs in several urban areas from 2011 to 2019. Considering the redistribution process of radiocesium on urban areas to WWTP mirroring that on river catchment areas, the availability of these data allowed studying the possibilities of using radiocesium concentrations in sewage sludge to evaluate the values of rapid and slow wash off rate. In this study, the temporal trend of radiocesium concentrations in sewage sludge was analyzed and the values of its rapid and slow wash off from the surface of corresponding urban areas were evaluated.

2. Methodology

2.1. Data Collection
Four urban areas were chosen for this study; namely Fukushima, Koriyama, Nihonmatsu and Miharu. As shown in Figure 1, radionuclide deposition, ^{137}Cs and ^{134}Cs concentration in sewage sludge and WWTP operational data from each of the urban areas were collected [2,7,8]. The WWTP in each urban area used the same technology which is activated sludge for biological process unit and sludge concentrator for sludge treatment unit. The service area of the four WWTP covers areas of 5,264, 8,070, 643, and 624 Ha, respectively serving a total of 550,000 inhabitants [7]. The four urban areas were categorized in green zone indicating that the area received about 150,000 to 1,000,000 Bq of radiocesium after the accident [8].

2.2. Temporal Analysis
The general model we used in this study was adapted from Smith et al (2004) which employ three phases of wash off rate as described in the equation Model 3 (Figure 1). c(t) (Bq/kg) is the concentration of

radionuclide in sewer sludge, D (Bq) is the initial deposition, k_1 and k_2 are the fast and slow wash off rate, respectively, and A, B, and C are the empirical coefficient representing the contribution of spontaneous, fast and slow transfer, respectively. Based on Model 3, we developed an additional two models. Model 1 describes the wash off phenomenon only consisting of one phase whereas Model 2 describes the wash off phenomenon consisting of two phases.

2.3. Parameter Quantification
The value of each model parameter was determined by the Least Square method. The method was run by Sigma Plot software resulting in each value of k_1, k_2, A, B, and C. The running must result in a convergent iteration to produce valid parameter values. We evaluated the three models by using the determination coefficient and standard error and chose the best fit model to be used for temporal analysis of radiocesium concentrations in sewage sludge.
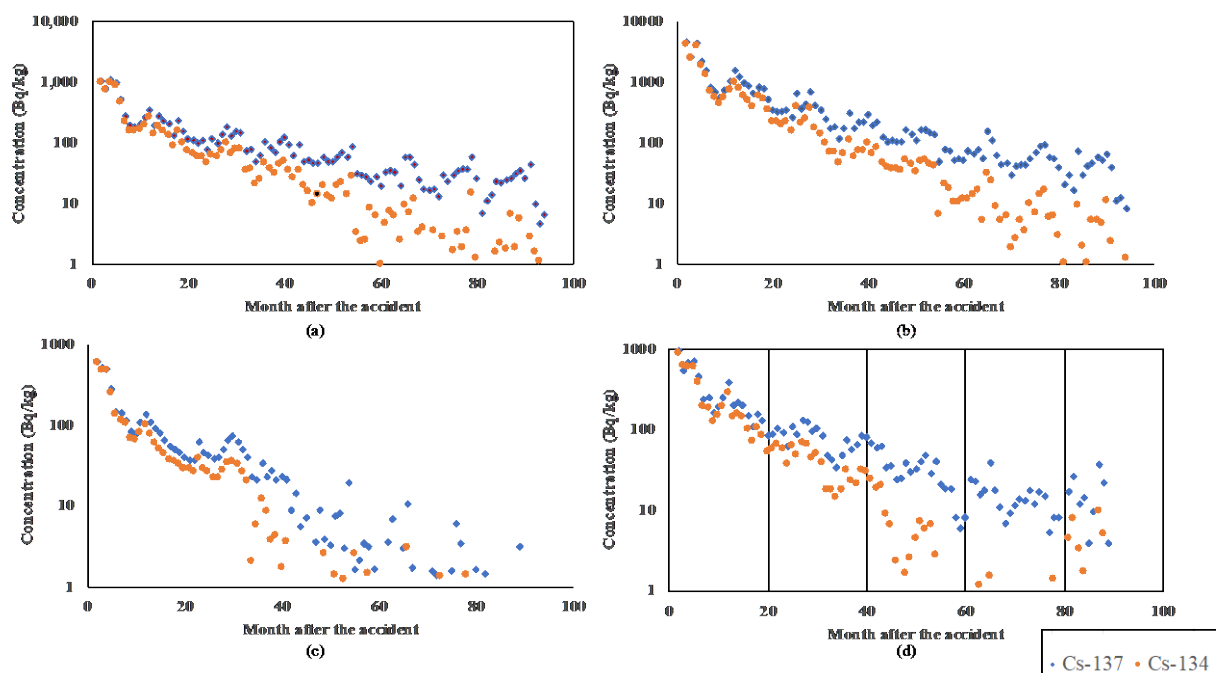

Figure 2. Radiocesium (^{137}Cs and ^{134}Cs) concentrations in sludge taken from WWTP in Fukushima, Koriyama, Nihonmatsu, and Miharu from May 2011 to March 2019

3. Results and Discussion

3.1. Time series of ^{137}Cs and ^{134}Cs concentration in sewage sludge
We used data provided by the local prefectural government of Fukushima which measures daily radiocesium (^{137}Cs and ^{134}Cs) concentrations in sludge taken from WWTP in Fukushima, Koriyama, Nihonmatsu, and Miharu in nine years period (2011-2019)[2]. The time-series data of the concentrations are shown in Figure 1. As can be seen, in the first two years after the accident, the concentrations were exceeded the safety limit set by the authority (100 Bq/kg). Concentrations of ^{134}Cs decreased faster than that of ^{137}Cs due to the half-life value of ^{134}Cs is only 2 years compared to that of ^{137}Cs (30 years).

3.2. Model Evaluation
Table 1 shows the values of determination coefficient and standard error for the fits of model Equation 1, 2, and 3 applied to the radiocesium concentrations in sewage sludge data. It can be seen that the model applying Equation 1 (consisting of spontaneous, rapid, and slow wash off) to the measurement data could not find a significant fit (NSF). The spontaneous wash off could not be detected in the

measurement data because the monitoring activity conducted by the local prefectural government started in May 2011. As suggested by Sassina (2008), the spontaneous wash off occurs on days or a few weeks after the first deposition[9]. The FDNPP accident happened in March 2011 whereas the first monitoring was conducted in May 2011, it is understandable that the model using Equation 1 could not find a significant fit to the measurement data.

Table 1. Coefficients of determination (R^2) of the different models to measurements of radiocesium concentrations in sewage sludge from four cities in nine years period.

Model	Fukushima		Kooriyama		Nihonmatsu		Miharu	
	^{134}Cs	^{137}Cs	^{134}Cs	^{137}Cs	^{134}Cs	^{137}Cs	^{134}Cs	^{137}Cs
Equation 1	NSF	NSF	NSF	NSF	NSF	NSF	NSF	NSF
Equation 2	0.895	0.825	0.881	0.71	0.935	0.872	0.946	0.909
Equation 3	0.911	0.892	0.911	0.888	0.955	0.922	0.963	0.955

The model applying Equation 2 (consisting of a single wash off rate) to the measurement data gave some satisfactory results. We obtained the values of the standard error of estimate in the range of 0.0004-0.0036 and 0.0002-0.0027 for ^{137}Cs and ^{134}Cs, respectively, indicating that the model predicts well. Having suggested that, it is quite doubtful that the time-dependent radiocesium flux from the surface of urban area to sewage sludge over nine years only has a single wash off rate. It contradicted several reports from Smith et al (2000) and Garcia-Sanchez (2008) which suggested that the radiocesium wash off fluxes are time-dependent and over a long period the fluxes flow due to two phases of wash off, rapid and slow process [3,4].

Among the three models, the best fit was obtained by applying Equation 3 (consisting of double wash off rates) to the measurement data. The values of the standard error of estimate were slightly smaller than those of the single wash off rate model (0.0003-0.0029 for ^{137}Cs and 0.0002-0.0024 for ^{134}Cs) indicating that the double wash off rates model predicted the measurement data better. As seen in Table 1, in all cities and both ^{137}Cs and ^{134}Cs, the R^2 values obtained from applying the double wash off rate model are closer to 1 than the values obtained by applying the single wash off rate model.

3.3. Wash off parameter values
Table 2 shows the values of parameters in the double wash off rates model obtained by the least square method embed in sigma plot software. As can be seen, the value of rapid and slow wash off rate is relatively constant over the four cities. The values of K_2 for ^{134}Cs are smaller than those for ^{137}Cs since the half-life of the radionuclide is only 2 years. The values of parameters A and B in Koriyama are significantly larger than the other three cities. This is because the WWTP in Koriyama city receives not only domestic wastewater flows but also stormwater inflows. Pratama et al (2018) estimated that only about 0.1% of radiocesium washed off on the surface of the urban area infiltrating the separated sewage system [9]. For a combined system, however, the percentage must be higher because it is designed to receive the stormwater inflows which contain a larger amount of radiocesium.

Table 2. Parameter values of the double exponential model

Model Parameters	^{134}Cs				^{137}Cs			
	Fukushima	Kooriyama	Nihonmatsu	Miharu	Fukushima	Kooriyama	Nihonmatsu	Miharu
A (kg/m^2)	0.0136	0.0796	0.0064	0.0129	0.0136	0.0812	0.006	0.013
k_1 (/month)	0.2356	0.3633	0.3157	0.3421	0.2411	0.389	0.2814	0.3512
B (kg/m^2)	0.0013	0.0113	0.0016	0.001	0.0019	0.0154	0.0015	0.0014
k_2 (/month)	0.0391	0.0597	0.0677	0.0546	0.0261	0.0468	0.0417	0.044

Conclusion

Based on the values of k_1 and k_2 obtained in this study, it was estimated that the rapid wash off occurred during the first 6-7 months after FDNPP accident whereas slow wash off occurred from the second year after the accident. The values of k_1 and k_2 compared to those reported in the river catchment scale after the Chernobyl accident is not significantly different. According to those reported values compiled by Garcia-Sanchez (2008), rapid wash off rates were in the range of 22-23 month while the range of slow wash off rates were 123-213 month. The results of this study suggest that the temporal trend of radiocesium concentrations gave important information on the wash off rate of the radionuclide on the surface of urban areas. Also, the rates reflect the natural decontamination process on a scale of the surface of a catchment area. Using the results of this study, our future work would be spatial analysis on the values of the model parameters. If we were able to clarify spatial factors that significantly affect the model parameters, we can build a general model that is relatively easy to predict concentration of radiocesium concentration in sewage sludge, in case another nuclear accident occurs.

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