Modeling Transport Behavior of Radiocesium through River Systems

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Abstract

This paper outlines circumstances regarding development of models to evaluate radioactive cesium behavior in river systems and discusses issues to be considered. The study used a relatively simple approach, parameterizing the transfer rate of radionuclides from one component to others, using, for example, soil-loss models and multi-compartment models. These models were applied to calculate the annually averaged transport behavior of radioactive cesium across entire river basins. On the other hand, detailed river models and water transport models were applied to evaluate the behavior during each high water event. A comparison of these models indicates that the amounts of discharged radioactive cesium brought mostly by wash loads are more or less successfully calculated by all models investigated. Liquid wash-off and/or desorption behavior is expected to be modeled in detail to calculate the future behavior of dissolved cesium, which is thought to migrate easily into ecosystems.

Key words: radioactive cesium, river system, transport modeling

1. Introduction

The Fukushima Daiichi Nuclear Power Station (FDNPS) accident caused the release of various radionuclides into the environment. These radionuclides were transported by wind and deposited directly or by rainfall, contaminating a wide area mainly in Fukushima Prefecture (Katata *et al*., 2012). Among these radionuclides, radioactive cesium isotopes (Cs-134, Cs-137) dominate external exposure and are important nuclides to address for radiation dose reduction. Decontamination work to reduce external exposure doses is ongoing in areas with larger amounts of radioactive cesium deposition and higher air dose rates, mainly in the living sphere. Over 70 percent of the radioactive cesium deposited in Fukushima Prefecture fell onto forested areas (Kitamura *et al*., 2014). Since most of the forests are located far from the living sphere, they are not planned specifically for decontamination, meaning radioactive cesium will remain there in the future.

Radioactive cesium remaining in forests is thought to move through the environment with natural forces such as rain and wind, eventually reaching the living sphere and the ocean. The Japan Atomic Energy Agency (JAEA) has investigated the transport behavior of radioactive cesium in the river systems of Fukushima Prefecture through continuous observation and laboratory experiments. In order to analyze their behavior to date and evaluate their future behavior and changes in radiation doses, various computer simulation tools have been developed based on different concepts and approaches. The present paper outlines circumstances regarding development of models to evaluate radioactive cesium behavior in river systems and discusses issues to be considered.

2. Modeling Approach

The behavior of radioactive cesium in the environment results from a varied combination of physical and chemical phenomena such as dispersion and advection in the ground, transport of particles by water flow and adsorption/desorption of mineral particles and nuclides. For modeling these processes, two approaches can be considered. One is an empirical approach in which the averaged migration velocity is evaluated based on various observed data, and the other is a phenomenological approach in which mathematical models expressing physical/ chemical phenomena are combined to predict behavior.

Calculating soil loss from terrestrial areas followed by calculating river transport is a typical empirical approach. Soil transport fluxes in each mesh point of the terrestrial area are calculated using a soil loss model and fluxes from the connecting mesh to the river are treated as inputs to the river at this point. This modeling approach is relatively simple and adequate for annually averaged behavior. It should be noted, however, that this approach assumes strong attachment of contaminants to soil particles in the terrestrial area. In the case of evaluating total Cs in the environment of Fukushima Prefecture, this assumption seems acceptable.

The phenomenological approach is based on the physical processes of water transport itself in the terrestrial area considering processes caused by surface water, for example, impact by rain drops, erosion by surface water, transport of soil particles by river water, and so on. This type of model can treat solid and liquid phases separately so that the behavior of adsorbed and dissolved cesium can be treated separately. It also considers not only simple instantaneous equilibrium but also kinetically controlled interaction between these phases. Such a physical model is consequently complicated and needs more calculation resources, but it has the benefit that the impact of single high water events like typhoons can be estimated, taking magnitude into account.

3. Models and Their Application

3.1 Soil Loss Analysis

The Universal Soil Loss Equation (USLE) is one of the basic models used in soil loss prediction. It is an empirical statistical model that considers planar erosion and rill erosion processes. Yamaguchi *et al*. (2014) developed the Soil and Cesium Transport (SACT) model, which can evaluate annually averaged transport of radioactive cesium together with soil loss based on the USLE. The model calculates 1) the possible amount of soil loss, 2) empirically predicted soil loss due to water flow, 3) soil loss among cells depending on slope, and 4) cesium transportation together with soil loss, in that order. In particular, the cesium concentration's dependence on soil particle size and cesium depth profile in the soil can be taken into account.

A sensitivity analysis was also carried out for some parameters, such as rainfall and runoff factors, critical shear stress of erosion, grain size distribution and flow velocity in lakes. As a result, the critical shear stress of erosion showed the highest sensitivity to the amount of soil erosion, while other parameters showed less sensitivity (Yamaguchi *et al*., 2014). The annual flux of Cs-137 from the Abukuma River to the ocean during the first year after the accident was estimated at 3.0 TBq/y with 0.23 – 3.7 variation in the sensitivity analysis. This value agrees well with 5.34 TBq/y which was reported as the Cs-137 flux from Aug. 2011 to May 2012 (Yamashiki *et al*., 2014). The calculated fluxes of other rivers running through the coastal area also showed relatively good agreement with observations (Kitamura *et al*., 2014).

In these calculations, it was pointed out that some of the assumptions might cause discrepancies between calculations and observations. The first criticism was that the initial deposition of radioactive cesium on water surfaces like rivers, lakes and ponds was ignored, since they only occupied less than 2 percent of the targeted area. The second criticism was that the river flow rate averaged over the past 10 years was used. The USLE itself is an empirical approach and it is essential that it use statistically evaluated parameters, so fluxes for years with larger high water events seem to be underestimated. The third criticism was that the depth profile of radioactive cesium in the topsoil was considered stable. Matsuda *et al*. (2015) reported that the relaxation depth β (g cm^{-2}) was found to increase by an average of 30 percent over approximately one year, based on depth profiles annually obtained using the scraper plate method at more than 80 locations. This indicates that radioactive cesium has downwardly migrated in the soil with time. If the depth profile is assumed constant, the amount of cesium discharged together with soil loss is overestimated and residual cesium in the soil is conversely underestimated. To be precise, the depth profile of radioactive cesium showed relatively high sensitivity regarding the discharge of radioactive cesium in the sensitivity analysis, in which a 58 percent smaller relaxation depth α (m^{-1}) gave a 53.6 percent smaller annual flux of radioactive cesium from the Abukuma River Basin (Yamaguchi *et al*., 2014). This can be easily explained by decreased soil surface concentrations, which are related to the total deposition by the following equation:

$$C_0 = R_A \alpha \tag{1}$$

where C_0 is the activity concentration of radioactive cesium (Bq m^{-3}) and R_A is the radioactive cesium inventory (Bq m^{-2}).

3.2 Multi-Compartment Model

Pratama *et al*. (2015) developed a multi-compartment model, which incorporated the process of radioactive cesium transport from the ground surface into the rivers of the Abukuma River Basin. In this model, compartments in the environment, such as trees, litter, topsoil, deep soil, etc., were designated as storage points of radioactive cesium, and transfer rates among each compartment were defined empirically or based on mechanistic models. Calculated monthly radioactive cesium flux values at the mouth of the Abukuma River showed good agreement with those observed by Yamashiki *et al*. (2014). In addition, the upper reaches of Abukuma River were divided into 29 small catchments, and the relationship between radioactive cesium flux and land use was determined in each catchment. As a result, the ratio of forested area showed a negative relationship to the flux and that of urban area, a positive one, indicating that less radioactive cesium was discharged from forests but more from urban areas. Consequently, the lower Abukuma reaches, which are occupied less by forests and more by urban areas, showed larger discharges of cesium than the upper reaches.

On the other hand, two sources of uncertainty were pointed out. One was failure to consider the effect of the size of suspended particles on transportation behavior, and the other was applicability of the wash-off coefficient obtained from post-accident Chernobyl studies. The flux from the Abukuma River evaluated with the multi-compartment model was larger than that with the SACT model. As mentioned above, however, the flux of the first year was close to the observed flux. This can be explained by the fact not only was soil wash-off considered, but also liquid wash-off, which was not taken into account in the SACT model but small enough to be disregarded, particularly in the first year. It has also been suggested that the coefficients used in the multi-compartment model were mostly acceptable, at least in the evaluation during the first year after the accident.

3.3 One-Dimensional River Transport Model

Phenomenological models were used for transport analysis of radioactive cesium in river systems. Kitamura *et al*. (2015) applied the Time-Dependent One-Dimensional Degradation and Migration (TODAM) code to the Ukedo River System in combination with the SACT model, the calculation results of which were used as input data. In this code, five fractions of cesium are considered; dissolved, adsorbed in sediments, and adsorbed in suspended particles, which are divided into three size fractions. The basic equations involved advection-dispersion, sedimentation and re-floating, inflow/outflow of each fraction, and kinetically controlled sorption and desorption equilibrium of cesium into sediments and suspended particles. The results showed that bed contamination is mostly dominated by sediment erosion and deposition rather than sorption and desorption of cesium in the bed. Most of the cesium migrated in a dissolved form under low flow rate conditions, while particulate cesium dominated migration under high flow rate conditions.

Kurikami *et al*. (2014) analyzed the inflow and outflow behavior of radioactive cesium in the Ogaki Dam reservoir located in the middle reaches of the Ukedo River using TODAM. Focusing on a typhoon rainfall event in September 2013, the observed flow rate and inflow concentrations of dissolved and particulate cesium were used as input data, and outflow behaviors were evaluated. The calculation results showed good agreement with observations.

3.4 One-Dimensional Water Transport Model

Kinouchi *et al*. (2015) proposed a phenomenological model. They based it on the Water and Energy Transfer Process (WEP) model, which could take into account physical water movements such as evaporation, evapotranspiration, infiltration, surface runoff, sub-surface runoff, groundwater flow, overland flow and river water flow. A model that could deal with erosion induced by raindrop impacts and hydraulic erosion was additionally coupled. It should be noted that this model did not take the size effect of soil particles into account. Adsorption equilibrium between soil particles and radioactive cesium was also ignored and it was assumed that radioactive cesium was strongly adsorbed in soil particles.

This model was applied to evaluate the river flow rate and concentration of suspended matter during high water events in the Kuchibuto River System. The calculation results showed good agreement with the observed values. On the other hand, in spite of conservatively ignoring the adsorption equilibrium of radioactive cesium, the concentration of radioactive cesium in suspended matter tended to be underestimated compared to observed values, especially in July 2011 and December 2012. These discrepancies could be explained by the lacking factor of higher mobility of radioactive cesium deposited on the forest canopy and in thawing water, in July 2011 and December 2012, respectively.

3.5 Two-Dimensional River Transport and Sedimentation Model

Determining the distribution of cesium accumulating on flood plains is essential to evaluating the effect on air dose rates in the living sphere near rivers. For this purpose, what is needed is a two-dimensional transport model which can consider cross-sectional variation of river transport characteristics. Iwasaki *et al*. (2015) analyzed the deposition behavior of radioactive cesium near the Abukuma River estuary using a two-dimensional river water transport model. This model considered two-dimensional advection and dispersion, sedimentation and resuspension. The concentration of cesium in sediment particles was fixed. Analysis of a high water event from a typhoon in September 2011 showed that most of the radioactive cesium transported as wash load directly reached the sea, and only a small amount accumulated on the flood plain or river bed in the decreasing stage of the hydrograph.

Analysis of the sedimentation behavior of the Ogaki Dam reservoir using a similar model was also reported (Yamada *et al*., 2015). The calculation results showed the behavior of each fraction of cesium as follows: the whole sand fraction accumulated near the inflow to the reservoir, while the silt fraction reached the outlet with part of it flowing out downstream. Almost all of the clay fraction flowed out downstream.

3.6 Two-Dimensional Water Transport Model

A two-dimensional water transport model involving both surface and subsurface water flows was developed (Mori *et al*., 2015). This model could consider transport of sediment induced by raindrops and water flow, and transport of radionuclides in both dissolved and particulate fractions. Solid and liquid phase radionuclide distributions were considered in kinetically controlled equilibrium interactions. The model was applied to the Hokkawa Dam reservoir basin in Fukushima Prefecture, and the water and sediment discharge into the reservoir was reasonably reproduced. A sensitivity analysis of the distribution coefficient was also carried out. The simulated cesium concentrations in the reservoir bottom sediment with distribution coefficients of $4\text{x}10^5$ and $5\text{x}10^6$ L/kg were similar to measured values, while the results of a simulation with $1.2\text{x}10^3$ L/kg were lower.

The same model, GETFLOWS, was applied to simulate the discharge of cesium from six river systems in the coastal area (Sakuma *et al*., 2017). The ratio of discharge at each heavy rain fall event in 2013 to the total amount of cesium deposited in the basin was calculated and the total discharge ratio of all events in 2013 varied from 0.07 to 0.33 percent.

4. Model Summary and Discussion

The results of calculating cesium discharge from some of the river basins are compared in Table 1. Among these models, the multi-compartment model tends to overestimate, and water transport models such as WEP and GETFLOWS tend to underestimate the discharge. As mentioned above, one of the uncertainties of the multi-compartment model was its use of the wash-off coefficient obtained from post-accident Chernobyl studies. Therefore, optimization of parameters using data obtained in Fukushima's environment is expected to give more reasonable results. In the cases of WEP and GETFLOWS, the results for rivers without dams, such as the Kuchibuto and Kuma rivers, seem reasonable; while for those with dams, such as the Ukedo and Tomioka rivers, they seem to underestimate it. Their estimation of the amount of radioactive cesium discharged from the upper mountainous area was reasonable, but those from lower areas were underestimated.

The fine fraction of discharged soil was considered to dominate the transport of radioactive cesium in the lower reaches of rivers with dams due to the sedimentation effect of the dams, while the coarse fraction dominated in rivers without dams. Kinouchi *et al*. (2015) introduced no classification of soil particle size in the calculations by the WEP model, so the larger reference particle size given as mean particle size resulted in underestimation of the discharge. The result of GETFLOWS was the total amount discharged during nine high water events in 2013 (Sakuma *et al*., 2017). Radioactive cesium was transported as a fine or dissolved fraction at low flow rates. In river systems without dams, radioactive cesium discharge was characterized by a relatively higher

Table 1 Comparison of characterization and calculation results of transport models.

(a)Abukuma River

Model / Observed	Depth distribution	Size effects	Discharge flux	Duration
SACT*1	Exponential	3 sizes	3.0 TBq/y	1st year
Multi-compartment*2	3 layers	No	6.5 TBq/y	1st year
Observed*3			4.8 TBq/y	2011.3–2012.3

b)Kuchibuto River

Model / Observed	Depth distribution	Size effects	Discharge flux	Duration
Multi-compartment*2	3 layers	No	2.1 GBq/d	1st year
WEP*4	Exponential	No	0.75 GBq/d	2011.9–2012.12
Observed*3			0.69 GBq/d	2011.3–2012.3

(c)Rivers in the coastal area

Model	Depth distribution	Size effects	Discharge flux	Duration
SACT*5	Exponential	3 sizes	2.0 TBq/y (Ukedo)	1st year
			0.28 TBq/y (Kuma)	1st year
			0.11 TBq/y (Tomioka)	1st year
GETFLOWS*6	All grids	All fractions	0.6 TBq (Ukedo)	2013 1)
			0.20 TBq (Kuma)	2013 1)
			0.060 TBq (Tomioka)	2013 1)
Observed*7			1.0 TBq/y (Ukedo)	2013 2)

References: *1 Yamaguchi *et al*. (2014), *2 Pratama *et al*. (2015), *3 Yamashiki *et al*. (2014), *4 Kinouochi *et al*. (2015), *5 Kitamura *et al*. (2014), *6 Sakuma *et al*. (2017), *7 JAEA (2014).

1) Total discharge of nine high water events in 2013.

2) The amounts discharged observed for the Ukedo and Takase rivers from Oct. 2012 to Oct. 2014 were added and normalized to annual flux.

proportion of discharge at low flow rates. Therefore, it was possible to obtain more realistic values with GETFLOWS if the discharge at low flow rates was taken into account. Thus, the uncertainty of the fine fraction's behavior may have caused the discrepancy in radioactive cesium discharge in the river systems with dams.

Several computer simulation codes were compared from the viewpoint of dimensionality and consideration of water flows, sediments and radionuclide transport (Mori *et al*., 2015). It should be noted that similar results could be obtained by these models, especially in the upper reaches of the river basin, in spite of differences in their treatment of features. This means that most of the amount of radioactive cesium transported could be expressed by relatively simple processes such as soil loss.

Since dissolved cesium is highly mobile and easily taken into ecosystems, predicting its behavior is highly desirable. For this purpose, the generation processes and their formulation and parameterization should be defined based on observed results. Liquid wash-off and/ or desorption are considered to play an important role in the processes, which occur not only in surface water but also groundwater, especially at points where groundwater flows out. Models that can take these processes and groundwater flow into account are desired to confirm their contribution.

5. Concluding Remarks

Various types of models were developed to calculate the transport behavior of deposited cesium in the environment. It became possible to predict the behavior of radioactive cesium throughout whole river basins during various rainfall events. To verify the applicability of models, it is essential to compare their calculation results with observed values and optimize their parameters based on measured values. Model calculations of the fate of radioactive cesium in the environment in the future are expected to be used as basic information in managing forestry, agricultural and fishery activities.

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