An Integrated Watershed Modeling to Assess the Long-term Fate of Fukushima-Derived Radionuclides

Koji Mori¹, Kazuhiro Tada¹, Yasuhiro Tawara¹, Koichi Ohno², Mari Asami², Koji Kosaka² and Hiroyuki Tosaka³
¹Geosphere Environmental Technology, Corp., Japan
²National Institute of Public Health, Japan
³Department of Systems Innovation, The University of Tokyo, Japan
¹mori@getc.co.jp

Abstract: The Fukushima Dai-Ichi Nuclear Power Plant (NPP) accident was induced by the Great East Japan Earthquake and the subsequent Tsunami on March 11th, 2011. Radionuclides (RNs) which emitted into the atmosphere from the NPP fell onto the land surface by precipitation. The fallout RNs were then redistributed through surface/subsurface water flow and sediment transport. To study this redistribution phenomena, we developed a novel integrated watershed modeling tool to assess the long-term fate and transport of the fallout RNs and their environmental impact. The feature of the simulator is to compute fully-coupled behavior of RNs transport among channels, overland and subsurface using physics-based 3D numerical model. An actual reservoir basin was selected as a study area and the measured concentration was reproduced to obtain a better understanding of the radionuclide behavior. Reasonable matching between the simulated $^{137}$Cs concentration and the observed one was achieved. The computed results showed that suspended sediment was the dominant transport media of $^{137}$Cs, and its fraction of the total $^{137}$Cs flux was more than 95%. Furthermore, it was indicated that the total discharge of $^{137}$Cs by surface water flow and sediment transport was less than 5% of the initial inventory, and most of the deposited $^{137}$Cs was still remaining within the watershed. These quantitative results and visualized contents using GIS shall contribute to effective discussion on planning of regional decontamination and reconstruction.

Keywords: Watershed modeling; Radionesium; Fukushima Dai-Ichi Nuclear Power Plant; Sediment transport; GETFLOWS.

1. INTRODUCTION

Radionuclides (RNs) emitted into the atmosphere during the Fukushima Dai-ichi Nuclear Power Plant (NPP) accident fell onto the land surface over a wide area with rainfall or in the form of aerosol. After that, the fallout RNs were redistributed through surface/subsurface water flows and sediment transport. The fate and transport of RNs are affected by regional watershed conditions, such as meteorology, land use/land cover (LULC), topography, surface geology, and so forth. After the NPP accident, the Japanese Government and international cooperative organizations carried out decontamination to reduce the impact of radiation exposure on public health. There should be some limits to capture the whole picture of RNs redistribution even if a number of monitoring stations were installed in the field, and it is necessary to apply numerical simulation for deep understanding and future predictions. Since the exchange of surface and subsurface contaminated water is concerned depending on regional specific conditions, surface and subsurface coupling become relevant in the numerical simulation. Moreover, because some RNs can adsorb onto the surface of sediment particles, the redistribution induced by both water and suspended sediment need to be modelled as multiple transport media. These comprehensive modelling makes it possible to predict a real time RNs redistribution and a long-term spatiotemporal variation. To develop an efficient decontamination and/or reconstruction plan with their appropriate interpretation, such simulated results become helpful. Furthermore, predicting the future give us more valuable information for early warning in emergency situations (i.e. RNs discharge pathways during heavy rainfall, etc.) and for prioritization of various reconstruction projects. Meanwhile, most of the existing codes cannot take into account RNs transport in watershed-scale problems. Limited computer codes that can simulate the RNs behavior with sediment transport were
developed after the Chernobyl NPP accident in 1986. The RNs transport codes RIVTOX (Zheleznyak, 2003), TODAM (Onishi et al., 2013) and GSTAR1D (USDOI, 2006) are used for one dimensional river network system. THREETOX (Margvelashvily, 1996) is a three dimensional surface water modeling system for hydrodynamics, sediment and RNs transport in lakes, reservoirs, estuaries and coastal ocean regions. All of these codes cover only partial components of a watershed, such as rivers or specific bodies of water, and it is impossible to apply them to the entire watershed. Therefore, this study has aimed to develop a fully integrated watershed modeling technique for simulating the long-term fate and transport of fallout RNs. In order to validate the developed modeling technique, we applied this to a real reservoir basin and tried to reproduce the radiocesium concentration in the reservoir. In this paper, we provide a brief overview of the development model and demonstrate its applicability to an actual field.

2. RADIONUCLIDE TRANSPORT IN SURFACE AND SUBSURFACE COUPLED FLUID FLOW

2.1 Conceptual model

Figure 1 shows a schematic view of fallout RNs redistribution in the watershed system. RNs deposited on the land surface can be transported by surface water flow, subsurface water flow and sediment transport. There are two major transport media for RNs: water (aqueous phase) and suspended sediment (solid phase). Coupled surface and subsurface water flows become a background flow field for RNs redistribution. Whereas sediment particles with adsorbed RNs can be transported through surface water flows. These different RNs species discharge into rivers, lakes and oceans through both surface water and groundwater in the model. Although contaminated sediment particles can infiltrate and travel through subsurface porous media, it is considered to be limited in RNs redistribution because the clogging effect reduces the sediment mobility. Therefore, we assume such colloid transport process by groundwater flow is neglected for the simplicity. Moreover, in order to assess the fate of both short- and long-lived RNs and its proper impact to water quality in reservoir, we included radioactive decay during redistribution in the model. The spatial distribution of meteorology, topography and LULC in the watershed are key factors that can affect RNs redistribution. These land surface heterogeneity was taken into account in the developed model.

![Figure 1. Schematic of the fallout radionuclide redistribution in watershed](image)

2.2 Governing equations

The system of governing equations is expressed by fluid flow, sediment and RNs transport coupled processes. Fluid flow is modeled as air/water two phase, surface and subsurface coupled fluid flows,
and the interactive behavior of the suspended sediment and RNs in flow field is considered. The governing equations are comprised of the mass conservation laws of fluid flow, suspended sediment particles in surface water, water dissolved RNs, RNs absorbed onto suspended sediment particles and RNs in deposited sediment particles, as described below.

\[ \nabla \cdot (u_w - q_w) = \frac{\partial (\phi S_w)}{\partial t} \]

(1)

\[ \nabla \cdot (u_g - q_g) = \frac{\partial (\phi S_g)}{\partial t} \]

(2)

\[ \nabla \cdot (u_w R_{s,i}) + \nabla \cdot \varepsilon_{s,i} \nabla R_{s,i} - q_w R_{s,i} - f_{s,i} - f_{e} = \frac{\partial (\phi S_w R_{s,i})}{\partial t} \]

(3)

\[ \nabla \cdot (u_g R_{a,i}) + \nabla \cdot \varepsilon_{a,i} \nabla (R_{a,i} R_{a,i,j}) - q_w R_{a,i,j} + \lambda R_{a,i,j - 1} + z_{a,i,j} e_{a,i,j}^{w} = \frac{\partial (\phi S_w R_{a,i,j})}{\partial t} \]

(4)

\[ \nabla \cdot (u_g R_{s,i}) + \nabla \cdot \varepsilon_{s,i} \nabla (R_{s,i} R_{a,i,j}) - q_w R_{s,i,j} + \lambda_{s} R_{s,i,j - 1} + z_{s,i,j} e_{s,i,j}^{w} = \frac{\partial (\phi S_w R_{s,i,j})}{\partial t} \]

(5)

\[ - \frac{1}{1 - \phi} \sum (a f_{s,i}^{w} + a f_{e,i}^{w}) = \frac{\partial \xi}{\partial t} \]

(6)

where \( u \) is the flow velocity (m/s), \( q \) is the volumetric flux of the sink and source (m\(^3\)/m\(^3\)/s), \( \phi \) is the effective porosity (m\(^3\)/m\(^3\)), \( S \) is the fluid saturation (m\(^3\)/m\(^3\)), \( R_s \) is the volume of suspended sediment per unit volume of surface water (m\(^3\)/m\(^3\)), \( \varepsilon_{s,i} \) is the turbulent diffusion coefficient (m\(^2\)/s), \( f_{s,i}^{w} \) is the soil detachment rate by raindrop impact (m\(^3\)/m\(^3\)/s), \( f_{e}^{w} \) is the rate of soil erosion/deposition due to surface water flow (m\(^3\)/m\(^3\)/s), \( R_a \) is the volume of dissolved RNs per unit volume of water (m\(^3\)/m\(^3\)), \( D_{ij} \) is the dispersion coefficient (m\(^2\)/s), \( \lambda \) is the decay constant of RNs (1/s), \( z_{a,i,j}^{ads} \) is the volume flux for RNs due to adsorption and desorption (m\(^3\)/m\(^3\)/s), \( R_{s,i,j}^{ads} \) is the volume of adsorbed RNs per unit volume of suspended sediment (m\(^3\)/m\(^3\)), \( a \) is the water depth (m) and \( \xi \) is the elevation of the bed surface (m). Subscript \( w \), \text{g}, \text{i} and \text{j} denote water phase, gas phase, grain size of sediment particle and radionuclide species, respectively. \( \nabla \) is the differential operator and \( t \) is time (s).

The Manning’s law for surface water and the generalized Darcy’s law for subsurface fluid flow are respectively used to compose the flow term of these equations. A depth-averaged diffusive wave approximation of the Saint-Venant equation is employed for surface water flow. Since there is no porous material above land surface, the effective porosity in all equations can be set to 1.0. The detail formulation of surface and subsurface coupling is in Tosaka et al. (2000, 2010). The sink and source terms of sediment transport \( f_{s,i}^{w} \) and \( f_{e}^{w} \) can be evaluated by selecting the suitable models from the existing different models for both cohesive and non-cohesive materials.

The detachment of soil particles by raindrop requires kinetic energy. The amount of kinetic energy that reaches the bed surface varies depending on LULC. When the surface is covered by water body, the kinetic energy of raindrops is absorbed by both water and soil, and soil detachment is affected. Torri et al. (1987) took into account the reduction effect of kinetic energy of water by the following exponential.

\[ f_{s,i}^{w} = k_s E_{r,i} e^{-bh} \]

(7)

where \( k_s \) is the Soil Detachability Index (SDI) of a soil mixture (kg/J), \( E_{r,i} \) is the kinetic energy of raindrops per unit area of the bed surface for each sediment particle \( i \) (J/m\(^2\)/S), and \( b \) is an empirically related parameter related to the texture of the bed surface. We assumed that every particle of the bed surface material is equally exposed to rain, and that the kinetic energy of raindrops is consumed in proportion to the fraction of each grain size. The kinetic energy \( E_{r,i} \) of each raindrop is expressed by the weighted form \( E_{r,i} = a_i E_{r} \), where \( a_i \) is the fraction of the amount of particles with grain size \( i \) to the total amount of bed surface sediment.

The SDI is defined as the weight of soil particles detached per unit of rainfall energy. In simple terms, the SDI can be calculated by dividing the total detachment by the total energy of the rainfall. Morgan et al. (1998) showed the typical value of SDI is 1.4–2.4 (g/J) for clay, 0.8–2.3 (g/J) for silt and silty loam and 1.0–6.4 for sand and sandy loam (g/J). The SDI is affected by the soil moisture content of the bed surface. Although SDI of the soil mixture varies depending on many environmental conditions, we simply assumed that the effective SDI can be described by a linear function of water saturation.

On hillslopes, erosion and deposition induced by water flow are often described using the parameter of transport capacity, which represents the equilibrium sediment load (detachment and deposition) that can be transported by overland flow. Many different expressions using simple flow hydraulics (such as stream power, unit stream power and shear stress) have been proposed for modeling erosion and deposition.
deposition induced by water flow. Depending on the erosion process (rill, inter-rill or sheet erosion), various transport capacity models can be easily incorporated in the governing equations. Assuming that all particles of the bed surface are equally exposed to water flow, the conventional transport capacity for each sediment particle \( i \) can be expanded as follows (Morgan et al., 1998):

\[
f_{s,i}^c = \beta_s \rho_{s,i} \overline{V}_{s,i} (TC - C_{ss,i})
\]

Here, \( \beta_s \) is a correction factor for the cohesion of bed material (dimensionless), \( C_{ss,i} \) is the volume concentration of sediment particles defined by the particle volume per unit volume of surface water (\( \text{m}^3/\text{m}^3 \)), \( \overline{V}_{s,i} \) is the settling velocity (\( \text{m/s} \)), and \( TC \) is the transport capacity. The correction factor \( \beta_s \) takes into account the cohesion dependence of the flow bed erodibility, that is, \( \beta_s \) is 1 for non-cohesive materials and between 0 and 1 for cohesive materials. Kabir et al. (2011) proposed the generalized power law \( \beta_s = A \eta^B \) using the empirically derived parameters \( A \) and \( B \), which should be determined from the site conditions, where \( \eta \) (kPa) is the cohesive strength. For simplicity, we adopted Rubey’s modified equation for the settling velocity of each sediment particle (Rubey, 1933).

### 2.3 Discretization and solution procedure

The governing equations were spatially discretized using an integral finite difference method. A fully implicit temporal discretization was employed to achieve stable computation. Due to the strong non-linearity of fluid properties, the Newton-Raphson method is employed to ensure that the result converges. In each time step, surface and subsurface coupled fluid flow simulation is conducted firstly to determine the water velocity field, which includes exchanges between surface and subsurface fluid flows. Then, using the computed velocity, the advection term in Equation (3) is composed for sediment transport simulation. Suspected sediment transport by advection and turbulence diffusion is computed in surface water. After the other advection terms in Equations (4) and (5) are composed for different RNs species, total simulation of RNs transport is performed using the computed water and suspended sediment velocity fields. All of these processes have been incorporated into the GETFLOW simulator (e.g. Tosaka et al. 2000, 2010; Mori et al. 2011).

### 3. APPLICATION TO THE FUKUSHIMA-DERIVED \(^{137}\)Cs REDISTRIBUTION

#### 3.1 3D numerical modeling of fallout radiocesium in the reservoir basin

There are several multi-purpose reservoirs (i.e. irrigation, drinking and flood prevention, etc.) around the Fukushima Dai-Ichi NPP. A typical reservoir basin that has role of the regional drinking water supply was selected as the study area. There were no particular reasons to be emphasized in this site selection. The basin is located at the southern part of Fukushima Prefecture. The distance from the Fukushima Dai-Ichi NPP is approximately 90 km. Figure 2 shows the topographical relief within the reservoir basin with an area of 15 km\(^2\). The rock-filled dam within the basin completed in 2000 for flood prevention and drinking water supply. The total reservoir capacity is 5,500,000 m\(^3\), and the flooded area is 37 ha. The water supplies to neighboring communities in the lower area of the dam site, and the maximum daily water supply is more than 20,000 m\(^3\). Annual mean precipitation is approximately 1,900 mm. Land surface within the watershed is widely covered by forest, and the subsurface geology is predominantly comprised of pyroclastic sedimentary.

Figure 3 shows the developed 3D grid-block system of the reservoir basin. The horizontal spatial resolution of the grid-blocks was approximately 20-50 m. In the vertical direction, the subsurface geology was discretized into 27 layers varying in thickness, and the total number of grid-block was 990,711. The daily precipitation and air temperature data were used as the distributed forcing data to land surface. Using the Harmon method (Harmon, 1961), the annual mean evapotranspiration was estimated to be 400 mm. The LULC classification is used to determine the interception of precipitation and the surface roughnesses. The hydraulic parameters were assigned to each grid-block based on the geological classification. Table 1 shows the permeability and the effective porosity used for each of them, which includes the parameters calibrated using hydrographs based on measured data. Rainfall-induced soil detachment rate \( f_{s,\text{DP}}^c \) is evaluated using raindrop kinetic energy and soil detachability based on Torri et al. (1987). Transport capacity for rill erosion is applied to evaluate the rate of soil erosion/deposition by water flow (Morgan et al. 1998). The surface soil in the study area is comprised of Andsol, Brown earth and Podsol, and was modeled as with multiple grain diameter of \( 10^{-5}, 10^{-3} \) and \( 10^{-4} \) m, which roughly correspond to clay, silt and fine sand respectively. We assumed that the initial...
thickness of the contaminated surface soil layer was 2 cm. The composition of each grain size is assumed to be 25% of clay, 25% of silt and 50% of sand. The soil detachability is assumed to be 1.68 g/J as the initial guess based on the representative value from Morgan et al. (1998). Radiocesium $^{137}$Cs, which has a long half-life (30.07y), was selected for the fate simulation. It is well known that $^{137}$Cs is adsorbed strongly to sediment particles. We assumed here that $^{137}$Cs can adsorb to all of the fine particles.

![Figure 2. Study area with topographic relief of the reservoir basin](image)

![Figure 3. 3D grid-block system. Different color shows topographical elevation of land surface (left) and geological classifications (right)](image)

| Table 1. Hydraulic parameters for each geological unit |
|---------------------------------|-----------------|
| Permeability (m/s) | Effective porosity (%) |
| Surface soil | $10^{-5}$ (upper), $10^{-3}$ (lower) | 50 |
| River bed deposit | $10^{-4}$ (horizontal), $10^{-5}$ (vertical) | 30 |
| Piedmont sediment | $5.0 \times 10^{-5}$ | 30 |
| Debris avalanche sediment | $5.0 \times 10^{-5}$ | 30 |
| Sliding soil mass | $5.0 \times 10^{-5}$ | 30 |
| Nasu volcano-lava group | $10^{-5}$ (horizontal), $10^{-6}$ (vertical) | 20 |
| Dacite pyroclastic sediment | $10^{-6}$ | 20 |
| Weathered rock | $10^{-6}$ | 20 |
| Base rock | $10^{-8}$ | 10 |

Table 2 lists the transport parameters. Spatial distribution of the initial inventory of $^{137}$Cs was assumed to be homogeneous based on published data from MEXT (2014). The distribution coefficient Kd, which is defined as the ratio of the contaminant concentration on the solid phase to the contaminant concentration in the liquid phase at equilibrium state, is one of the most important parameters to affect RNS movement. Since Kd varies widely depending on the soil properties and the chemical environment, the sensitivity of $^{137}$Cs distribution on Kd was examined. As a general value of Kd, the average and maximum values for various soil types in IAEA (2010) are used in Case1 and Case2,
respectively. In addition, an identified Kd by Nagao et al. (2013), which was derived from measured concentration, is used in Case3. For tortuosity and dispersion coefficient of the subsurface soil, there were no available measured data. These parameters were determined from the general literatures.

<table>
<thead>
<tr>
<th>Initial inventory of $^{137}$Cs (Bq/m$^2$)</th>
<th>Case1</th>
<th>Case2</th>
<th>Case3</th>
</tr>
</thead>
<tbody>
<tr>
<td>Molecular diffusion coefficient (m/s)</td>
<td>$2\times10^{-9}$</td>
<td>$2\times10^{-9}$</td>
<td>$2\times10^{-9}$</td>
</tr>
<tr>
<td>Distribution coefficient (mL/g)</td>
<td>1,200</td>
<td>400,000</td>
<td>5,000,000</td>
</tr>
<tr>
<td>Tortuosity factor</td>
<td>0.5</td>
<td>0.5</td>
<td>0.5</td>
</tr>
<tr>
<td>Dispersion length (m)</td>
<td>0.1(L), 0.01(T)</td>
<td>0.1(L), 0.01(T)</td>
<td>0.1(L), 0.01(T)</td>
</tr>
</tbody>
</table>

a) MEXT, 2014; b) IAEA, 2010; c) Nagao et al, 2013; d)(L) longitudinal, (T) Transverse

Based on these conditions, the flow field in equilibrium state is firstly reproduced. Then transient response simulations during recent 3 years (from May 2011 to December 2013) are executed using daily-meteorologic forces, and the resultant change in surface soil thickness induced by sediment erosion and deposition was examined. After that, the simulated $^{137}$Cs concentration in the bottom sediment was extracted to compare with the measurement. Contaminated sediment was obtained from a depth of 10 cm in the lake bottom, and the concentration of $^{137}$Cs was measured. Measurement data in such aquatic environments are published by the Government of Japan (MEXT, 2014). Finally, $^{137}$Cs inventory maps were produced by subtracting the initial $^{137}$Cs inventory from the end state obtained in the simulation.

3.2 Results and discussions

Figure 4 shows a plane projection of the computed 3D streamlines in equilibrium state, which depicts the conjunctive surface and subsurface water flows using lines in different colors. From the computed spatial distribution, it can be seen that the groundwater which has been recharged in the upland areas discharges into the lower valley and maintains the base flow. Figure 5 shows the resultant change in surface soil thickness induced by sediment erosion and deposition. Positive values (red and orange) represent deposited regions, and negative ones (blue) represent eroded regions. The steep hillslopes adjacent to the rivers indicate major eroded regions.

Figure 6 shows the computed time evolution of suspended and deposited $^{137}$Cs concentration for different Kd values at location R3 (see Figure 2), which is inside of the reservoir. In the results, the breakthrough of $^{137}$Cs to the reservoir can be seen at the end of April, after that, the concentration fluctuates with the sudden increase in every typhoon season. The measurement data have shown a large variation within the range from 500 to 4,000 Bq/kg. The variation seems to be decreased with time. With the exception of Case 1, the concentration of $^{137}$Cs in the simulation results with high Kd values gradually increased. Although there is a large discrepancy between the measurement and simulation results, Case 2 and Case 3 were reproduced the lower level of the measured concentration. In Case 1, the $^{137}$Cs concentration was much lower than the measured concentration. From these results, it can be identified that the magnitude of Kd was $10^5$–$10^6$ mL/g at least. The root mean square error (RMSE) for all of the measured data was 1,702 Bq/kg for Case2 and 1,600 Bq/kg for Case3. In both cases, RMSE in each year were gradually decreased with time: 2,495 Bq/kg in 2011, 911 Bq/kg in 2012 and 232 Bq/kg in 2013 for Case3. These indicate that there were spatial heterogeneity of initial inventory of $^{137}$Cs, which was not considered in the model. Therefore, the computed and measured data is getting closer with time. The cumulative discharge of $^{137}$Cs after NPP accident was extracted from the computed results. According to that, the fraction of suspended $^{137}$Cs to the total discharge was more than 95%. This tendency is almost consistent with the observed data in different locations by MEXT. Also, most of the fallout $^{137}$Cs was still remaining within the watershed in the computation. The total discharge from the dam site to downstream was less than 5%.

Figure 7 shows the resultant spatial redistribution of the $^{137}$Cs inventory due to water flow, sediment and $^{137}$Cs transport in the watershed. Contaminated regions where the $^{137}$Cs inventory increased (decreased) from the beginning are given in red and orange (blue). Although the Kd is different in each case, the contaminated regions where the $^{137}$Cs inventory increased are identified in the primary rivers and the reservoir in all cases. Regions with decreased concentration are identified as the steep hillslopes near the primary rivers. The dominant $^{137}$Cs transport mechanisms are different, and specific redistribution patterns are formed depending on the Kd value. In Case 1, the amount of $^{137}$Cs in aqueous phase can be large, and therefore decontamination by water flow was faster compared with Cases 2 and 3. The spatial patterns of $^{137}$Cs redistribution in Cases 2 and 3 were almost identical.
Simulated streamlines of surface and subsurface conjunctive water flow

Figure 4.

Simulated spatial patterns of erosion and deposition (change in soil thickness)

Figure 5.

Simulated time evolution of suspended $^{137}$Cs concentration in the reservoir at R3 in Figure 2 (left) and deposited $^{137}$Cs concentration in bottom sediment (right).

Figure 6.

Simulated changes in spatial distribution of $^{137}$Cs inventory for different Kd values

Figure 7.
4. CONCLUSIONS

We developed a novel watershed modeling tool for simulating the fate and transport of fallout RNs. The developed technique enables the comprehensive simulation of RNs redistribution through surface and subsurface water flows and sediment transport coupled processes. Both water-dissolved RNs and ones adsorbed on suspended sediment are interactively transported with phase transfer between water and solid phases. To evaluate the applicability of the developed simulator, we applied this technique to an actual reservoir basin and simulated the fate and transport behavior of Fukushima-derived radionuclides for a period of recent three years after the NPP accident. The simulated $^{137}$Cs concentration in the reservoir bottom sediment were consistent with the latest measured data, and the spatiotemporal patterns of $^{137}$Cs redistribution were considered to be reproduced reasonably. In future work, we plan to improve the applicability of this modeling technique by using more information gathered through extensive environmental monitoring. Moreover, it can be used to support political decision making for efficient decontamination and reconstruction projects.

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