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# Integrated watershed modeling for simulation of spatiotemporal redistribution of post-fallout radionuclides: Application in radiocesium fate and transport processes derived from the Fukushima accidents

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A B S T R A C T

Simulation of the watershed-scale fate and transport of radionuclides is required in order to predict the consequences of contamination redistribution. Integrated watershed modeling is a suitable technique for this task, but it requires fully coupled investigation of radionuclide behavior in surface water, suspended sediment and subsurface aquifers. We developed a novel simulator for computing the spatiotemporal redistribution of fallout radionuclides in watersheds. The simulator was applied to an actual reservoir basin contaminated by fallout radionuclides from the Fukushima Dai-Ichi Nuclear Power Plant accident in 2011. As a result, the simulated $^{137}Cs$ concentration in bottom sediment showed a reasonably close match with the measurement data. The distribution coefficient of $^{137}Cs$ consistent with the latest measurement data was identified as being at least 400,000 L/kg, and it was estimated that more than 90% of the total $^{137}Cs$ distributed in the fallout remains in the catchment area.



## 1. Introduction

The interaction of fallout radioactive isotopes with the hydrologic cycle has been studied in detail to interpret water and sediment behavior. Environmental tracers such as $^{3}H$, $^{36}Cl$, $^{85}Kr$ and $^{14}C$ are used for estimating water recharged area or dating old groundwater (e.g., Balonov et al., 2010; IAEA, 2013). Radiocesium isotopes ($^{134}Cs$, $^{137}Cs$) derived from nuclear weapons testing in the 1950s have been used as typical tracers for sediment in watersheds since most of the radiocesium is adsorbed onto surficial sediment or organic matter (e.g., Golosov, 2000; Poreba, 2006; Meili and Worman, 1996). Only a limited amount of radiocesium remains dissolved in the aqueous phase due to its strong adsorption capacity (Yamaguchi et al., 2012; Owen et al., 1996). To verify the water flow and sediment transport behavior estimated using such environmental tracers, reactive solute transport simulation with the distributed load is often performed (e.g., Sanford, 2011; Shimada et al., 2012a, 2012b; Ichiyanagi et al., 2012).

Radionuclides emitted into the atmosphere during the Fukushima Dai-ichi Nuclear Power Plant (NPP) accident fell to the land surface over a wide area with rainfall or in the form of aerosol. After that, the fallout radionuclides were considered to be redistributed by surface/subsurface water flows and sediment transport. The fate and transport of the dissolved and adsorbed radionuclides are affected by regional watershed conditions, such as meteorology, land use/land cover (LULC), topography and surface geology. After the Fukushima Dai-ichi NPP accident, the Japanese Government and international cooperative organizations carried out decontamination to reduce the impact of radiation exposure on public health (e.g., Kitamura et al., 2014; IAEA, 2014). Surface soil removal is one of the typical temporary countermeasures, but the

*Abbreviations:* IAEA, International Atomic Energy Agency; JAEA, Japan Atomic Energy Agency; LULC, Land use and land cover; MEXT, Ministry of Education, Culture, Sports, Science and Technology, Japan; NPP, Nuclear power plant; SDI, Soil detachability index; U.S. DOI, U.S. Department of Interior Bureau of Reclamation.

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application of this technique for redistributed fallout is rather time-consuming. Although many ground-based or airborne-based measurements of radioactivity have been conducted by the Ministry of Education, Culture, Sports, Science and Technology (MEXT) of Japanese Government (MEXT, 2014), there are limitations to understanding the whole picture by measurements alone. To gain the deep understanding of the radionuclide transport necessary to develop an efficient decontamination plan, it is essential to conduct collaborative work with numerical simulation of radionuclide redistribution. Numerical simulation is a useful tool for predicting the long-term fate and transport of fallout radionuclides, and it allows us to evaluate the following: the length of the recovery period; whether there is any substantial groundwater and/or soil contamination; the amount of radionuclide discharge, including that resulting from radionuclide redistribution (e.g., infiltration to subsurface aquifers and discharge into rivers and ocean); and whether there are regional differences in terms of radionuclide contamination. To address these issues, it is important to model the distinctive phenomena governing the redistribution of fallout radionuclides that have been heterogeneously deposited over large catchment areas. Part of the deposited radionuclides runoff with surface water and suspended sediment, some of the aqueous species infiltrate into the subsurface and become adsorbed onto soil particles, depending on the adsorption capacity of the soil material. The infiltrated radionuclides can be transported by groundwater flow and subsequently discharged into rivers or lakes, eventually reaching the ocean.

Since the redistribution of fallout radionuclides in watersheds is a consequence of water flow and sediment transport in the surface and subsurface environment, coupling of both sets of data is relevant in numerical simulation. Fully coupled modeling makes it possible to predict radionuclide redistribution under a wide range of spatiotemporal variation (e.g., real-time and long-term forecasting). Appropriate interpretation of simulation results is therefore extremely useful in developing an efficient decontamination and/or reconstruction plan. Furthermore, future predictions give valuable information for early warning scenarios in emergency situations (i.e., radionuclide discharge pathways during heavy rainfall and so on) as well as for prioritization of various reconstruction projects.

Table 1 compares the dimensionality and capability of selected computer codes for surface/subsurface water flows, sediment and radionuclide transport modeling compared with the GEneral-purpose Terrestrial fluid FLOW Simulator GETFLOWS, which is discussed below. There are many physics-based computer codes form modeling watershed-scale sediment transport, such as EUROSEM (Morgan et al., 1998), CLEAMS (Kinsel and Nicks, 1980), KINEROS (Woolhiser et al., 1990), WEPP (Foster and Lane, 1987), MIKE-SHE (Graham and Butts, 2005; Danish Hydraulic Institute, 2007) and InHM (VanderKwaak, 1999; Heppner et al., 2006; Ran et al., 2007). These computer codes have been reviewed in detail, and their specific functions are summarized in the associated literature (e.g., Borah et al., 2003; Daniel et al., 2011; Heppner et al., 2006; Ran et al., 2007). Some comparative studies on soil erosion models using multiple codes have also been carried out (e.g., Centeri et al., 2009).

Because some radionuclides are strongly adsorbed onto the surface of sediment particles, both water and suspended sediment can serve as transport media. However, most of the existing codes cannot take into account radionuclide transport in watershed-scale problems. Limited computer codes that can simulate the radionuclide behavior with sediment transport were developed after the Chernobyl NPP accident in 1986. The radionuclide transport codes RIVTOX (Zheleznyak et al., 2003), TODAM (Onishi and Yokuda., 2013) and GSTAR1D (U.S, DOI, 2006) are used for one-dimensional river network systems. THREETOX (Margvelashvily et al., 1996; Zheleznyak, 1997) is a three-dimensional surface water modeling system for hydrodynamics, sediment and radionuclide transport in lakes, reservoirs, estuaries and coastal ocean regions. All of these codes cover only small parts of a watershed, such as rivers or specific bodies of water, and it is impossible to apply them to the entire watershed.

Under the widely fluctuating environmental conditions near the land surface, fallout radionuclides travel through various watershed components such as hillslopes, river channels, valleys and subsurface aquifers. To implement watershed modeling, most existing software codes require individual watershed components and their interconnections to be explicitly defined (i.e., disconnection and reconnection of watershed components). However, since water flows fluctuate continuously, they are difficult to

**Table 1**
Comparison of dimensionalities and capabilities of selected computer codes for surface/subsurface water flows, sediment and radionuclide transport simulation.

| Model name | Dimensionality | | Coupling scheme | Hydrology | | Sediment | Radionuclide | |
|---|---|---|---|---|---|---|---|---|
| | Surface | Subsurface | | Surface | Subsurface | Surface | Surface | Subsurface |
| EUROSEM[a)] | 1D | n/a | n/a | n/a | n/a | HS, C | n/a | n/a |
| KINEROS[b)] | 1D | 1D | DU | KW | AE | HS, C | n/a | n/a |
| WEPP[c)] | 1D | 1D | DU | KW | AE | HS, C | n/a | n/a |
| MIKE-SHE[d)] | 1D/2D | 1D/3D | DU | SV, DW | BE | HS, C | WD, SS | WD, AD |
| InHM[e)] | 2D | 3D | FC | DW | R | HS, C | WD | WD, AD |
| ParFlow[f)] | 1D | 3D | FC | KW | R | n/a | n/a | n/a |
| CATHY[g)] | 1D | 3D | FC | DW | R | n/a | n/a | n/a |
| SHETRAN v4[h)] | 1D/2D | 1D/3D | DU | SV, DW | BE | HS, C | WD, SS | WD, AD |
| MODHMS[i)] | 2D | 3D | FC/IC | DW | R | n/a | n/a | n/a |
| HydroGeoSphere[j)] | 2D | 3D | FC | DW | R | n/a | WD | WD, AD |
| RIVTOX[k)] | 1D | n/a | UC | DW | n/a | C | WD, SS | n/a |
| TODAM[l)] | 1D | n/a | UC | n/a | n/a | C | WD, SS | n/a |
| THREETOX[m)] | 3D | n/a | UC | NS | n/a | WB | WD, SS | n/a |
| GSTAR1D[n)] | 1D | n/a | UC | SV | n/a | C | n/a | n/a |
| GETFLOWS[o)] | 2D | 3D | FC | DW | GD | HS, C | WD, SS | WD, AD |

*.a) Morgan et al., 1998; b) Woolhiser et al., 1990; c) Laflen et al., 1991; d) Danish Hydraulic Institute, 2007; e) VanderKwaak, 1999; f) Kollet and Maxwell, 2006; g) Bixio et al., 2000; h) Ewen et al., 2000; i) Panday and Huyakorn, 2004; j) Therrien et al., 2010; k) Zheleznyak et al., 2003; l) Onishi and Yokuda, 2013; m) Margvelashvilii et al., 1996; n) U.S. DOI, 2006; o) Tosaka et al., 2000.

** 1D: one-dimensional; 2D: two-dimensional; 3D: three-dimensional; DU: Degenerated uncoupled; UC: Uncoupled; IC: Iterated coupled; FC: Fully coupled; KW: Kinematic wave; SV: Saint Vernant; DW: Diffusion wave: NS: Navier–Stokes equations; R: Richards equation; BE: Boussinesq equations; AE: Algebraic equations; GD: Generalized Darcy's law; HS: Hillslope; C: Channel/channel network; WB: Water body; WD: Water-dissolved species; SS: Suspended species; AD: Adsorbed species.