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Technical article

# Study on Radiation Dose Assessment in Contaminated Soil Due to Deposition of Suspended Radioactive Material

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**ABSTRACT** Radioactive materials dispersed in the atmosphere can be deposited in soil and have long-term radiological effects on the population depending on the physical properties of the material, but it is difficult to easily calculate the radiation dose of nuclides deposited in soil. In this study, based on previous studies, we used the PHITS code to evaluate the radiation dose with depth of deposited material in soil for $^{134}$Cs, $^{137}$Cs, $^{131}$I, $^{129\,m}$Te, $^{110m}$Ag, $^{238}$Pu, and $^{238+240}$Pu. Calculations were based on nuclide concentrations per unit area for contaminated soil at a radius of 10 metres from the ground surface. It was found that the contributions of $^{134}$Cs,$^{137}$Cs are very large, with the contribution of $^{134}$Cs initially being more than 70%, but with time, the contribution of $^{137}$Cs, which has a relatively longhalf-life, is large. It is possible to estimate the dose of radionuclides deposited in soil through computer simulation, which is considered to be effective in terms of radiation protection.

**Key words:** Atmospheric dispersion, Soil contamination, Soil deposition, Effective dose, Radiological disaster

## 1. Introduction

Radioactive material released and dispersed into the atmosphere as a result of an accident at a nuclear facility contains a variety of radionuclides. After being carried by the wind in the atmosphere, it can fall back to the ground and settle on buildings, soil, and people. Because people do not feel symptoms immediately upon being exposed to radiation, in some cases they may not realize they are contaminated and continue with their daily activities, further spreading the radioactive material beyond the contaminated area. However, the deposition of this contamination in the soil and its effects on the people living there can have long-term effects of exposure, depending on the nature of the radioactive material.

Various studies have been conducted on the effects of radionuclides on the human body during radiological emergencies, revealing effective dose conversion factors and relative surface contamination densities for different deposition pathways and methods depending on the surface material, location, and surface particles [1,2].

Since the Fukushima nuclear accident, various studies on soil deposition of diffuse radioactive materials have been conducted in Japan, including tracking nuclide sources through dose rates [2], spatial distribution of sedimentary radionuclides [3,4], and impact assessment through cesium profiles in soil [5].

Saito, K etal. measured the radioactive fallout released from the Fukushima Daiichi Nuclear Power Plant accident on three occasions to determine the radioactivity per unit area and the nuclides using a portable gamma-ray spectrometer, and based on these measurements, created inventory distribution maps of $^{134}$Cs, $^{137}$Cs, and $^{110\,m}$Ag [7].

One of the most important aspects of radiological emergency management is the ability to quickly and appropriately make judgments and take action to protect the public and emergency workers. A radiological incident assessment should consider all critical information available at any given time and should be an iterative and dynamic process to review the response as more detailed and complete information becomes available [8].

Therefore, it is necessary to estimate the dose value of radioactive materials deposited in the soil to the population. Since it is not easy to determine the radiation effects at different soil depths, we used a Monte Carlo-based computer simulation code

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to evaluate the radiation effects of radionuclides deposited at different soil depths on human health.

This study examined data on the soil deposition process of radioactive substances diffused from the atmosphere by reviewing papers published overseas. Based on the data on the behavior of deposited nuclides, the radiation dose to humans was evaluated using the PHITS code according to the depth of deposition of radioactive substances in the soil through measurements made during the 10-year period following the Fukushima nuclear power plant accident.

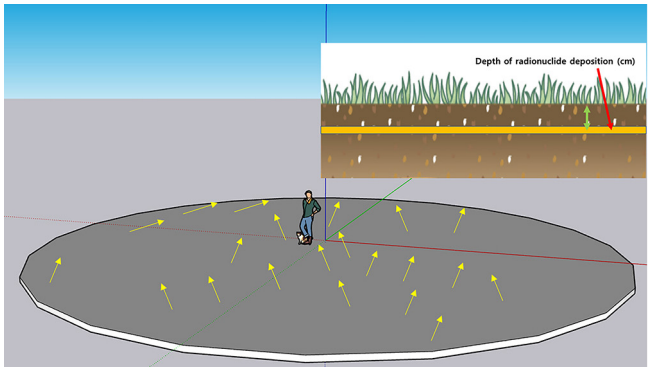


**Fig. 1.** External exposure of individuals to contaminated radioactive material.

## 2. Materials and Methods

### 2.1. Radiation Source Term

The accident at the Fukushima Daiichi Nuclear Power Plant resulted in the release of various radioactive materials. The types and distribution of radionuclides were investigated in this study to monitor the radiation risk to the population. The obtained data were used to calculate the radiation dose that individuals could receive from radioactive materials in contaminated soil.

Immediately after deposition, radionuclides deposited in the ground are assumed to form a planar source at the surface. Over time, these radionuclides migrate or leach into the soil, forming concentration depth profiles that vary depending on the form and shape of the soil. The distribution of radionuclides with respect to soil depth can be assumed to be due to an infinite number of planar sources in the ground, and these features can have different characteristics, resulting in peaks at different soil depths [9].

The spatial distribution and temporal changes of radioactive fallout released from the Fukushima Daiichi nuclear power plant accident were investigated through three sets of measurements carried out between early June 2011 and late 2012. The radioactivity per unit area of radionuclides with short half-lives such as $^{131}$I was measured using a portable gamma-ray survey meter. The radioactivity per unit area of radionuclides deposited in the ground soil was measured using a portable gamma-ray spectrometer. Soil samples taken in March, September, and December 2012 were mapped for $^{134}$Cs, $^{137}$Cs, $^{131}$I, $^{129m}$Te, and $^{110m}$Ag, which are important gamma-ray emitting nuclides that have the potential to expose residents to significant amounts of radiation in the early days after an accident [10].

The maximum deposited radioactivity was evaluated for $^{134}$Cs, $^{137}$Cs, $^{131}$I, $^{129m}$Te, $^{110m}$Ag, $^{238}$Pu, and $^{238+240}$Pu, as shown in Table 1.

### 2.2. PHITS

PHITS (Particle and Heavy Ion Transport Code System) is a Monte Carlo particle transport code developed by the Japan Atomic Energy Agency (JAEA). It statistically simulates the

**Table 1.** Spatial distribution of radioactive fallout from the Fukushima Daiichi nuclear accident (Saito K. *et al*. 2015)

| Nuclide | Maximum deposition density (Bq/m$^2$) | Distribution ratio (%) |
|---|---|---|
| $^{134}$Cs | $1.4\times10^{7}$ | 43.93436 |
| $^{137}$Cs | $1.5\times10^{7}$ | 47.07253 |
| $^{131}$[ | $5.5\times10^{4}$ | 0.17260 |
| $^{129m}$Te | $2.7\times10^{6}$ | 8.47306 |
| $^{110m}$Ag | $8.3\times10^{4}$ | 0.26047 |
| $^{89}$Sr | $2.2\times10^{4}$ | 0.06904 |
| $^{90}$Sr | $5.7\times10^{3}$ | 0.01789 |
| $^{238}$Pu | $4.0\times10^{0}$ | 0.00001 |
| $^{238+240}$Pu | $1.5\times10^{1}$ | 0.00005 |

**Table 2.** Mass ratio of the constituent substances of the soil [14]

| Nuclide | Mass ratio (g/cm$^3$) |
|---|---|
| H | 0.023 |
| C | 0.0023 |
| O | 1.22 |
| Na | 0.0368 |
| Mg | 0.005 |
| Al | 0.078 |
| Si | 0.775 |
| K | 0.0299 |
| Ca | 0.1 |
| Fe | 0.032 |

**Table 3.** Half-life and decay constant of the target nuclide [14]

| Nuclide | Half-life | Decay Constant (s$^{-1}$) |
|---|---|---|
| $^{134}$Cs | 2.07 y | 1.1. ×10$^7$ |
| $^{137}$Cs | 30.2 y | 7.2. ×10$^5$ |
| $^{131}$I | 8 d | 7.5. ×10$^3$ |
| $^{129m}$Te | 33.6 d | 1.8. ×10$^3$ |
| $^{110m}$Ag | 249.8 d | 2.4. ×10$^2$ |
| $^{89}$Sr | 50.7 d | 1.2. ×10$^3$ |
| $^{90}$Sr | 28.2 y | 7.7. ×10$^5$ |
| $^{238}$Pu | 87.7 y | 2.5. ×10$^5$ |
| $^{238+240}$pu | 6,563 y | 3.3. ×10$^3$ |

interaction of particles of various energies (neutrons, photons, charged particles, heavy ions, etc.) with matter, and is used in various fields such as radiation protection, nuclear fusion, and cosmic radiation [11,12].

This code is freely distributed and anyone can use and modify it. It also supports a wide range of particle types, including neutrons, photons, heavy ions, and more, so you can easily calculate events in particle behavior with ease. It also can model different forms of matter and complex systems. It provides highly accurate calculation results based on sophisticated physical models and utilizes multi-core processors to efficiently perform large-scale calculations [13].

Under the simulation conditions, the dose was calculated based on a unit area of contaminated soil at a radius of 10 meters from the ground surface. The density of the soil at the ground surface was 2.302 g/cm$^3$, which is the density value of soil provided by PNNL for the computational simulation, and the detailed composition was calculated as a mass fraction as shown in Table 2. The data for soil contamination was calculated using soil contamination concentrations from the data studied by Saito K. *et al*. [14].

Dose calculations were also performed at 3 cm and 5 cm to estimate dose values for radionuclides deposited in the soil as the average depth of deposition increased.

The Phits computer simulator has a calculating command called “tally”, which is a calculation command that references the ICRP’s recommended approach. To calculate the effective dose, the tally command in the PHITS code was used with the ICRP 103-based effective dose cards. The PHITS version was calculated using version 3.280.

### 2.3. Computer specifications

The computer used in the simulations was a laptop computer with a 13th Gen Intel (R) Core (TM) i5-1340P 1.90 GHz processor, 16.0 GB of RAM, and an operating system based on ×64.

## 3. Results and Discussion

Figure 2 shows the radiation dose values calculated by PHITS for a person at one meter above ground when the $^{137}$Cs nuclide is deposited in soil at a concentration of 1.5 × 10$^7$ Bq/m$^2$.

Table 4 shows the results of computer simulations of doses at a height of one meter above ground to assess the dose that

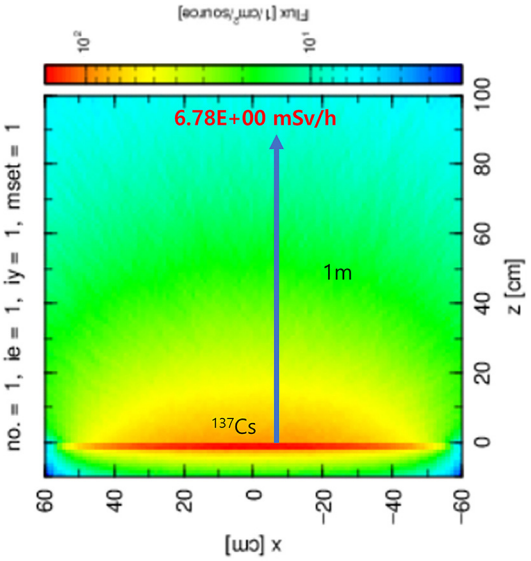


**Fig. 2.** Radiation dose at 1 m for $^{137}$Cs: Maximum deposition density 1.50×10$^7$ Bq/m$^2$

**Table 4.** Maximum deposited concentration of a radioisotope at a depth of 1 cm and the resulting dose at 1 m above ground surface

| Nuclide | Maximum deposition density (Bq/m$^2$) | Distribution fraction | Dose rate (mSv/h) | Relative error | Contribution percentage |
|---|---|---|---|---|---|
| $^{134}$Cs | $1.40\times10^{7}$ | 43.9% | $1.80\times10^{1}$ | 2.0% | 72% |
| $^{137}$Cs | $1.50\times10^{7}$ | 47.1% | $6.78\times10^{0}$ | 3.1% | 27% |
| $^{131}$I | $5.50\times10^{4}$ | 0.2% | $2.13\times10^{-2}$ | 3.0% | 0.1% |
| $^{129m}$Te | $2.70\times10^{6}$ | 8.5% | $1.70\times10^{-1}$ | 4.8% | 0.7% |
| $^{110m}$Ag | $8.30\times10^{4}$ | 0.3% | $2.44\times10^{-3}$ | 2.1% | 0.0% |
| $^{89}$Sr | $2.20\times10^{4}$ | 0.1% | $1.78\times10^{-6}$ | 1.8% | 0.0% |
| $^{90}$Sr | $5.70\times10^{3}$ | 0.0% | $6.57\times10^{-10}$ | 4.0% | 0.0% |
| $^{238}$Pu | $4.00\times10^{0}$ | 0.0% | $1.97\times10^{-9}$ | 4.1% | 0.0% |
| $^{238+240}$pu | $1.50\times10^{1}$ | 0.0% | $2.35\times10^{-5}$ | 2.6% | 0.0% |
| Sum | $3.19\times10^{7}$ | 100% | 2.50 ×10 | | 100% |

an occupant could receive from a contaminated ground surface. The higher dose rates for $^{134}$Cs and $^{137}$Cs at similar deposition concentrations are due to the fact that $^{134}$Cs having a shorter half-life than $^{137}$Cs, 2.065 years. However, the energy of the gamma rays emitted per decay is greater than that of $^{137}$Cs. This is because $^{137}$Cs emits 0.662 MeV of gamma rays per decay, whereas $^{134}$Cs emits 0.725 MeV of energy per decay.

Saito *et al*. (2015) reported that the contribution of the dominant radionuclide to the external effective dose could be converted into effective dose rate using a dose conversion factor that considered the average radionuclide deposition density of 50 representative sites. They found that $^{134}$Cs accounted for 28.1% while $^{137}$Cs accounted for 70.9%, which is likely due to the much longer half-life of $^{137}$Cs than $^{134}$Cs and its greater long-term effects.

Figure 3 shows the radiation dose distributions from PHITS' computationally simulated surface history for $^{134}$Cs, $^{137}$Cs, $^{131}$I, $^{129m}$Te, $^{110m}$ Ag, $^{238}$Pu, and $^{238+240}$Pu.

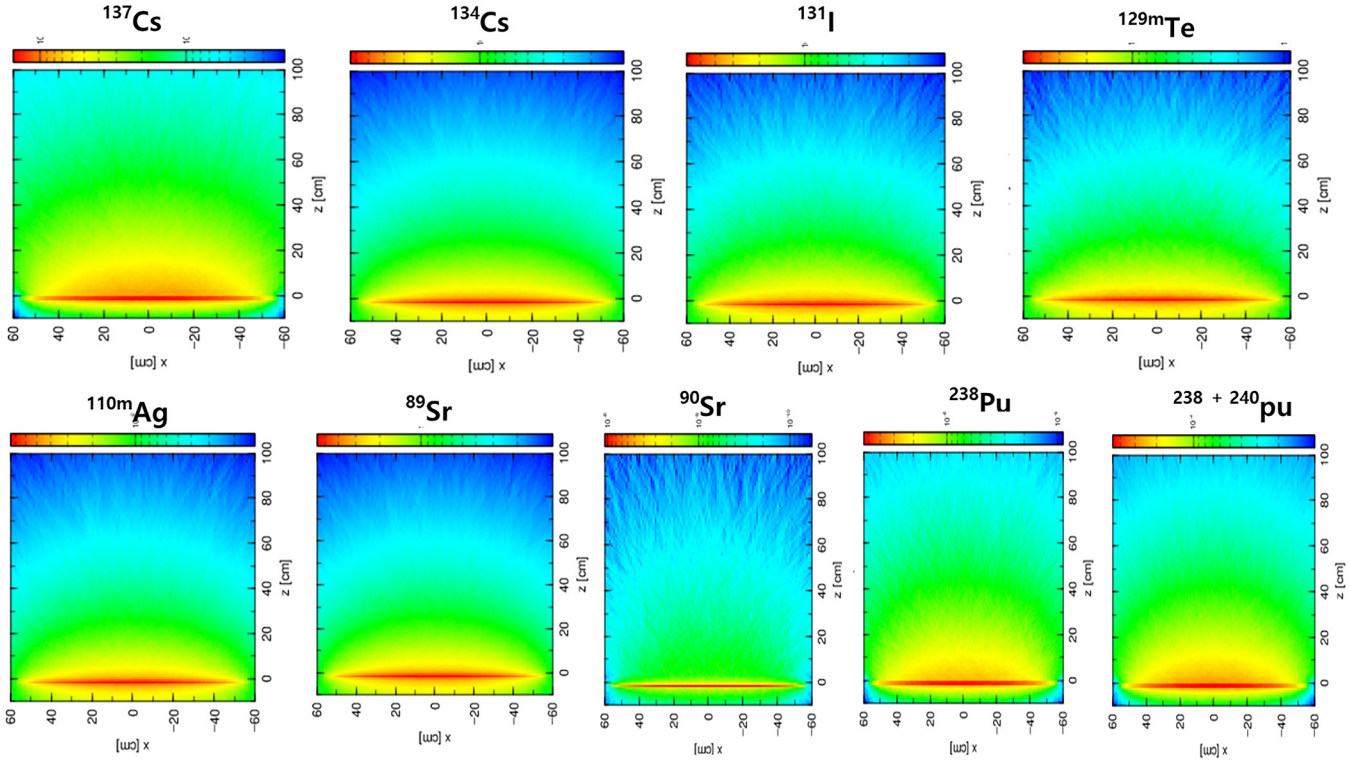


**Fig. 3.** Spatial dose distribution of radionuclide deposition at 1 cm depth in the ground

**Table 5.** Radiation dose and decay rate as a function of depth for nuclides

| Nuclide | Dose rate (mSv/h) at 1 cm | Dose rate (mSv/h) at 3 cm | Dose rate (mSv/h) 5 cm | Relative decay rate 3 cm | Relative decay rate 5 cm |
|---|---|---|---|---|---|
| $^{134}$Cs | $1.80\times10^{1}$ | $1.58\times10^{1}$ | $1.35\times10^{1}$ | 88% | 75% |
| $^{137}$Cs | $6.78\times10^{0}$ | $6.16\times10^{0}$ | $4.80\times10^{0}$ | 91% | 71% |
| $^{131}$I | $2.13\times10^{-2}$ | $1.48\times10^{-2}$ | $1.08\times10^{-2}$ | 69% | 51% |
| $^{129m}$Te | $1.70\times10^{-1}$ | $1.15\times10^{-1}$ | $9.05\times10^{-2}$ | 67% | 53% |
| $^{110m}$Ag | $2.44\times10^{-3}$ | $1.85\times10^{-3}$ | $1.46\times10^{-3}$ | 76% | 60% |
| $^{89}$Sr | $1.78\times10^{-6}$ | $1.38\times10^{-6}$ | $1.15\times10^{-6}$ | 78% | 65% |
| $^{90}$Sr | $6.57\times10^{-10}$ | $1.11\times10^{-10}$ | $1.08\times10^{-10}$ | 17% | 16% |
| $^{238}$Pu | $1.97\times10^{-9}$ | $1.97\times10^{-9}$ | $1.75\times10^{-9}$ | 100% | 88% |
| $^{238+240}$pu | $2.35\times10^{-5}$ | $1.93\times10^{-5}$ | $1.65\times10^{-5}$ | 82% | 70% |

**Table 6.** Effective dose to be received by residents: The dose coefficients presented in TECDOC-1162 (IAEA, 2000) were used to evaluate external and inhalation exposures

| Nuclide | Effective dose from deposition (mSv, 1 hr) | Effective dose from deposition (mSv, 1st month) | Effective dose from deposition (mSv, 2nd month) | Effective dose (mSv, 50 years) |
|---|---|---|---|---|
| $^{134}$Cs | $1.80\times10^{1}$ | $3.78\times10^{1}$ | $3.50\times10^{1}$ | $7.14\times10^{2}$ |
| $^{137}$Cs | $6.78\times10^{0}$ | $1.49\times10^{1}$ | $1.41\times10^{1}$ | $1.95\times10^{3}$ |
| $^{131}$I | $2.13\times10^{-2}$ | $1.38\times10^{-2}$ | $9.90\times10^{-4}$ | $1.49\times10^{-2}$ |
| $^{129m}$Te | $1.70\times10^{-1}$ | $2.97\times10^{-1}$ | $1.46\times10^{-1}$ | $5.94\times10^{-1}$ |
| $^{110m}$Ag | $2.44\times10^{-3}$ | $3.74\times10^{-1}$ | $3.24\times10^{-1}$ | $3.24\times10^{0}$ |
| $^{89}$Sr | $1.78\times10^{-6}$ | $2.42\times10^{-4}$ | $1.45\times10^{-4}$ | $6.16\times10^{-4}$ |
| $^{90}$Sr | $6.57\times10^{-10}$ | $9.69\times10^{-4}$ | $9.12\times10^{-4}$ | $1.20\times10^{-1}$ |
| $^{238}$Pu | $1.97\times10^{-9}$ | $1.56\times10^{-4}$ | $1.48\times10^{-4}$ | $2.64\times10^{-2}$ |
| $^{238+240}$pu | $2.35\times10^{-5}$ | $6.30\times10^{-4}$ | $6.00\times10^{-4}$ | $1.26\times10^{-1}$ |

Table 5 shows the computationally simulated dose values at deposition depths of 3 and 5 cm for each nuclide, compared to the values at the surface. It can be seen that the degree of attenuation depends on the nuclides emitting gamma rays and their energy.

In Table 6, the effective doses that residents will receive are evaluated for external and inhalation exposures using the dose factors presented in TECDOC-1162 (IAEA, 2000). Cumulative dose values can be calculated for the first month, second month, and 50 years after the accident. These values were evaluated for external and inhalation exposures by multiplying the maximum deposition density at the observation point by the dose conversion factor [15].

Radiation exposure from soil deposition of nuclides is initially dominated by $^{134}$Cs, which accounts for 70% of the total, but the long half-life of $^{137}$Cs results in a very large $^{137}$Cs deposition dose over a 50-year period.

In Table 6, all nuclides show a slight decrease in the dose after 2 months compared to the dose after 1 month, but since these are dose values accumulated over 50 years, they tend to increase.

## 4. Conclusion

Using soil sample data from the area around the Fukushima Daiichi nuclear power plant, we calculated the effective dose received by residents as a function of the surface deposition density of radionuclides using a PHITS computer simulation to calculate the spatial dose distribution. In particular, the graphical output capabilities of PHITS made it easy to view the quantitative values and visual shape distribution of the dose distribution. Effective dose values at the surface were calculated for each nuclide depending on the depth of deposition in the surface layer.

Although the committed effective dose values were obtained using separate dose conversion factors, we found that the ground dose distribution can be easily calculated from the post-incident ground surface concentrations. Furthermore, the dose distribution can be easily simulated computationally with the various nuclide data from PHITS, which facilitates dose prediction. The study confirmed that the radiation impact on the population due to nuclide deposition increases with the energy and half-life length of the nuclide.

Although it is assumed that atmospheric diffusion caused by a radioactive material accident occurs over flat terrain and the nuclide ratio is not constant because atmospheric conditions change over time, it was confirmed that the deposition concentration of each nuclide obtained through a general atmospheric diffusion simulation can be used to calculate the effective dose received by residents. In addition, it is difficult to calculate the regional characteristics of the radioactivity ratio uniformly due to the different deposition paths of radionuclides. Nevertheless, the value and visual distribution of the dose distribution can be easily obtained, and it is expected that this will expand the use of PHITS computer simulation.

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