

NUMERICAL SIMULATION FOR THE MOVEMENT OF CESIUM-137 IN GROUNDS

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ABSTRACT: Fukushima Dai-ichi nuclear disaster occurred by Great East Japan Earthquake in 2011. As the result, a lot of radioactive substances was discharged to atmosphere and deposited in surface of ground by rain. In damaged area, decontamination has been conducting. Usually decontamination for the ground is digging out for 5 cm from the surface of the ground, but its safety and validity is apprehended. In this study, permeation behavior analysis of radioactive substance in the ground was carried out in order to prepare new decontaminations. Specifically, behavior and advection-dispersion analysis was conducted by advection-dispersion equation was embedded a term that considered half-life. As a result, it is found that this analysis method has enough validity by comparing analysis result with result on the spot. Furthermore analytic evaluation for the current decontamination is performed.

Keywords: advection-diffusion analysis, infiltration, radioactive half-life, radioactive substances

1. INTRODUCTION

Tokyo an Electric Power Dai-ichi Fukushima Nuclear Power Plant accident has happened by the eastern Japan great earthquake in 2011. Therefore the radioactive substances released were carried by wind and fallen to the surface of the ground or the sea. Iodine-131, cesium-134 and cesium-137 have been detecting in the widespread land area such as the farmland and the ground of city area in Tohoku and Kanto [1].

Geo-Environmental pollution range by this accident was so huge that decontamination is continued now. Generally decontamination for the ground is digging and removing the ground layer about 5cm [2]. However analysis for behavior of radioactive substances has been ever examined. So the safety and validity for it is apprehended. Besides, the construction site of the radioactive material storage container becomes a problem. It is necessary to comprehend behavior of radioactive substances in soils in order to prepare new decontamination for the ground.

In experimental investigation handling radioactive substances is danger in experimental investigation. Therefore in this study, a term that considered radioactive half-life embedded to advection-dispersion equation in the order to evaluate infiltration behaviors of radioactive substances in the soils. The aim of this study is confirming validity for this analysis method by comparing analysis result with result on the spot. Furthermore examining various the soils have effects on infiltration behavior in the soils.

2. PRESENT CONDITION OF DECONTAMINATIONS FOR CONTAMINATED GROUNDS

In the decontamination guidelines, schoolyard, garden, park and farm land are designated as examination range of grounds decontamination. Generally, decontamination is conducted until air radiation is less than 0.23 mSv an hour [2].

For a procedure of decontaminating, at first, air radiation is measured before and after decontamination in order to confirm effect of decontaminating it. Next, decontamination is conducted at the hot spot, where the grounds is contaminated higher than the girth by storm drainage containing the radioactive substances, after covering around decontamination range with tarpaulin to prevent the radioactive substances spread. Some typical examples of the hot spot are cited as follows hollow, water pocket, ditch and spot under a tree. Still when an effect of decontamination isn't obtained sufficiently, coating of the grounds surface, reversal tillage and removing the grounds surface are selected and disturbed.

Coating of the grounds surface is a way to cover soils in the upper part, containing the radioactive substances with normal soils. Reversal tillage is a way to cover the grounds surface, which is approximately thickness 10cm, by replacing normal soils with soils of thickness 20cm containing the radioactive substances. It is found that an enough effect for removing the grounds surface is gained by removing depth approximately 5cm.

There are advantage in two former that air radiation would be fallen and the radioactive

substances would not be spread by cover, in addition, contaminated soils are not appeared. However, they are not radical contamination. Because the radioactive substances are existed in the grounds. Therefore, removing the grounds surface is disturbed with human power otherwise a heavy machinery in fact, like Fig. 1.

3. BEHAVIORS OF RADIOACTIVE SUBSTANCES IN GROUNDS

Radioactive substances fallen to the surface of the ground have four actions. They are concentration decrement with radioactive decay, moving in the soils, scattering and flowing out by weather and absorption by the plants [3].

3.1 Concentration Decrement with Radioactive Decay

Concentration decrement with radioactive decay depends on radioactive half-life. Radioactive half-life of Iodine-131 is about 8 days, its survival rate becomes 0% instantly. But radioactive half-life of cesium-137 is about 30 years so that long-term examination for it is necessary.

3.2 Movement in Grounds

3.2.1 Existence form of radioactive cesium in soils

The existence form of radioactive cesium and soil property and importance point moving in the soils. Radioactive cesium in the soils exists as monovalent cation, Cs^+ . There are clay minerals and organic matter which have a lot of negative electric charge on their surface. As with other heavy metals and positive ion, radioactive cesium adsorbs them.

3.2.2 Adsorbing with a cauterization agent

Most organic substance, which enter in the soils as animals and plants corpse like fallen leaf and branch, is disintegrate in water and carbon dioxide gas by soil animal and soil microbe. A part of them exits as difficulty degradability high molecular compound called cauterization agent in the soils. Cauterization agent have carboxyl, $-COOH$, on its surface. It has a negative charge by dissociating a hydrogen ion, H^+ . Consequently, Cauterization agent becomes adsorbent of radioactive cesium. Nevertheless, radioactive cesium which adsorbed with cauterization agent is permuted easily by other cation because this adsorbing is weak combination



Fig. 1 The decontamination for the ground

through the water around Cs^+ , hydrated water. Precisely, cauterization agent is adsorbent keeps the mobility of radioactive cesium is high. Consented radioactive cesium adsorbed with cauterization agent gradually transition to a selective high adsorbent in a process to repeat adsorption, desorption.

3.2.3 Adsorbing with a frayed edge site

2:1 type layer silicate, is one of the minerals constitute a clay particle, hold negative charges on silicon tetrahedral sheet. Expressly, mica holds a lot of negative charges then K^+ occupies six-membered ring. Consequently, other negative charges can enter into a layer because a layer interval already has been strongly closed.

Though, a distal portion of a layer swell in the state that kept seat structure by aeration. As a result, K^+ is released from a layer interval of a circumference part then hydration positive ion is hold in a layer interval. Wedge-shaped border part of the swelling layer and the non-swelling layer is called frayed edge site. In frayed edge site, hydration positive ion is released by spatial limitation. Then Cs^+ , whose hydration energy is the lowest moreover whose shape is fit for six-membered ring, adsorb strongly with frayed edge site, such as Fig. 2.

3.3 Scattering and Flowing out by Weather

As mentioned above, radioactive cesium adsorbs with clay mineral and organic substance then tends to take root on the spot. Besides it is found that adsorbed radioactive substance don't melt in water easily [4]. For that reason, there are no incident that radioactive cesium is detected from surface water and grounds water. However, There are researches that radioactive cesium moved very small [5]-[7].

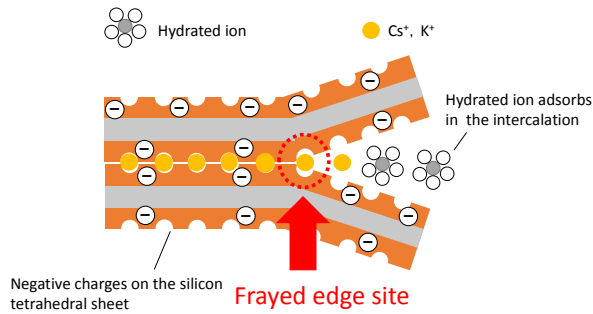


Fig. 2 Edge structure of a weathered mica

3.4 Absorption by Plants

There are two courses of the radioactive substances which were released transfer into plants. One is a course of the radioactive substances transfer into the surface of plant directly from atmosphere, called directly composed course. Another is a course of the radioactive substances which fallen grounds are absorbed from the root of plant, called root absorption course.

After Fukushima Daiichi nuclear power plant accident, directly composed course has a significant influence when quantity of descent of the radioactive substances is large. After it, root absorption course becomes influential. Furthermore, because pollution to plants by directly composed course depends on quantity of descent of the radioactive substances, the period is relatively short. On the other hand, most radioactive substances remain in the soils a long time due to them absorb with soils strongly. Therefore, pollution to plants by root absorption course lasts several decades because of radioactive half-life.

Quantity of absorption by root absorption course depends on concentration of the radioactive substances and transition coefficient. Transition coefficient, which is a parameter of tendency of the radioactive substances transfer into plants from soils, is equal to concentration of radioactive substance in edible part in a plant divided by concentration of radioactive substance in the soils.

Transition coefficient is difference from kind of crops. Average value of polished rice is 0.0005 and potato is 0.011 [8], [9].

4. INFILTRATION AND ADVECTION-DISPERSION ANALYSIS FOR RADIOACTIVE SUBSTANCES

Although scattering and flowing out by weather and absorption by the plants are extremely small values

and in order to evaluate pure infiltration behavior in the soils, in this study, density decrement with radioactive decay and moving in the soils are focused on.

Generally, substances moving in the soils analysis is modeled by the advection-dispersion equation. By this equation is added a term which considered radioactive half-life, it is applicable to moving analysis for radioactive substances.

4.1 Infiltration Equation

The general differential equation for two-dimensional infiltration can be expressed as Eq. (1).

$$\frac{\partial}{\partial x_i} \left(k \frac{\partial H}{\partial x_i} \right) + Q = \frac{\partial \theta}{\partial t} \quad (1)$$

Where: H = the total head, k = the hydraulic conductivity, Q = the applied boundary flux, θ = the volumetric water content, and t = time.

4.2 Advection-Dispersion Equation

The advection-dispersion equation which including radioactive half-life can be written as Eq. (2).

$$\theta \frac{\partial C}{\partial t} + \rho_d \frac{\partial S}{\partial C} \frac{\partial C}{\partial t} = \frac{\partial C}{\partial x_i} \left(\theta D_{ij} \frac{\partial C}{\partial x_j} \right) - U_i \frac{\partial C}{\partial x_i} - \lambda (\theta C - S \rho_d) \quad (2)$$

Where C is the concentration of radioactive substance, ρ_d is the dry mass density of the soils, D_{ij} is the dispersion tensor, U_i is the D'arcian velocity, λ is the decay coefficient and S is the adsorption.

The advection-dispersion equation is added a term with the decay coefficient λ that expresses the concentration decrement by radioactive half-life. It is leads to Eq. (3).

$$\lambda = \frac{\ln(2.0)}{T_{1/2}} \quad (3)$$

Where: $T_{1/2}$ is the radioactive half-life.

The Adsorption S is calculated by the linear adsorption isotherm with the distribution coefficient K_d , as shown in Eq. (4).

$$S = K_d C \quad (4)$$

Table 1 Parameters for silt

| | |
|----------------------------------------------------------------|----------------------|
| the hydraulic conductivity k (m/sec) | 1.0×10^{-6} |
| the volumetric water content θ | 0.4 |
| the dry mass density of the soils ρ_d (g/m ³) | 1.2×10^6 |
| the distribution coefficient K_d (g/m ³) | 8.0×10^{-3} |

Table 2 Parameters for Cs-137

| | |
|--------------------------------------------------------------------|----------------------|
| the radioactive half-life $T_{1/2}$ (years) | 30.2 |
| the coefficient of molecular diffusion D^* (m ² /day) | 7.2×10^{-6} |
| the initial concentration C_0 (Bq/m ²) | 3.0×10^8 |
| the dispersivity of porous medium α (m) | 0.01 |

Where K_d is the distribution coefficient.

The distribution coefficient K_d is index to express hydrophobicity and the transitivity of the chemical substance. This index is determined by the radioactive substance and soils moreover it is a slope of the linear adsorption isotherm.

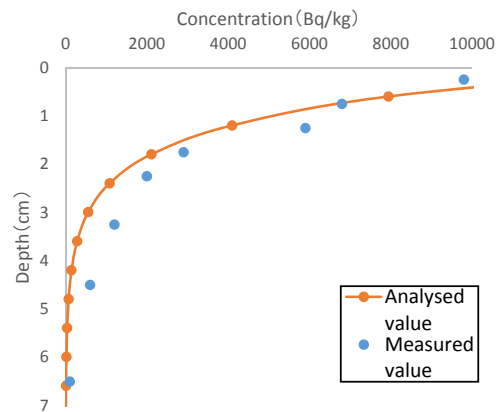
5. VALIDITY FOR THE INFILTRATION AND ADVECTION-DISPERSION ANALYSIS

5.1 Measured Results

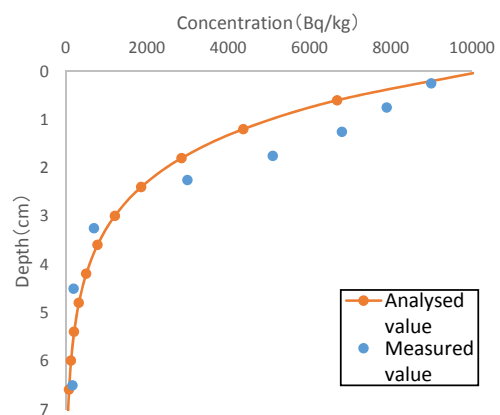
The analysis result of concentration distribution for Cs-137 are compared with measurement results in the area struck by Tokyo An electric power Daiichi Fukushima nuclear power plant accident in order to validate appropriateness of this analysis method. The measurement results was measured by Pref. Saito in the spot approximately 70km away from the Daiichi Fukushima nuclear power plant three times, 9 months progress, 18 months progress and 20 months progress [10]. According to the result, as depth becomes big, concentration becomes the tendency to be small exponentially. Besides Cs-137 had stayed by about 5 cm a long period of time. Besides by soils size measurement at the same time, the soils of the spot was proved to be the silt layer.

5.2 Conditions for the Analysis

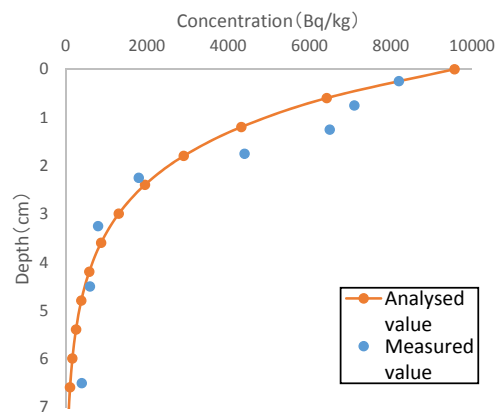
The analysis soils is set to the silt layer. The representative figures of each parameter of the silt is shown to Table 1 [11], [12]. On the other hand, about the radioactive substances, the coefficient of molecular diffusion and the initial concentration of



(a) 9 months progress



(b) 18 months progress



(c) 20 months progress

Fig. 3 The measurement result and analysis result of Cs-137 concentration in the depth direction

Cs-137 in this point was reported in the other report are used, as shown to Table 2 [13]. The analysis is entered these values and carried out.

5.3 Validity for the Analysis

Figure 3 shows the measurement result and analysis result of Cs-137 concentration in the depth direction.

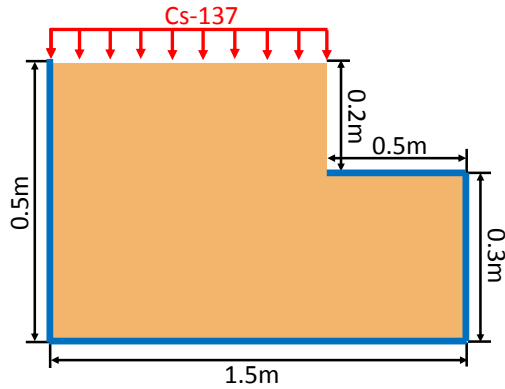


Fig. 4 The section for analysis

Table 3 Parameters for sand and clay

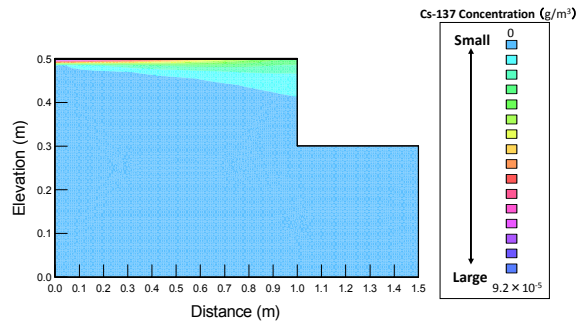
| | sand | clay |
|----------------------------------------------------------------|----------------------|----------------------|
| the hydraulic conductivity k (m/sec) | 1.0×10^{-5} | 1.0×10^{-7} |
| the volumetric water content θ | 0.4 | 0.4 |
| the dry mass density of the soils ρ_d (g/m ³) | 1.5×10^6 | 9.5×10^5 |
| the distribution coefficient K_d (g/m ³) | 4.0×10^{-3} | 1.0×10^{-2} |

According to it, their values are very closely in every depth and the tendency to decrease of analysis result is exponentially, like measurement result. Furthermore, they are the same even if time passes. Thereby the infiltration and advection-dispersion analysis including radioactive half-life has comfortable validity for the evaluation of infiltration behavior of radioactive substances in the soils.

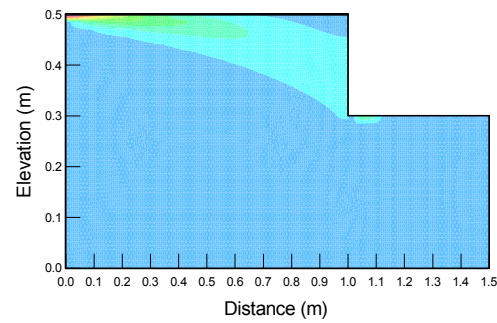
6. ANALYTIC EVALUATION OF CURRENT DECONTAMINATION

6.1 Conditions for the Analysis

The infiltration and advection-dispersion analysis including radioactive half-life is carried out in two cases, sand layer and clay layer, on the analysis section with water head difference which is descended Cs-137 which is necessary for long-term examination, like Fig. 4. Analysis section is set to the saturation state. And the boundary colored blue indicates the drainage condition. The parameters for sand and clay which necessary for the analysis are used their central values showed Table 3 [11], [12]. About parameters of Cs-137, they are entered the same as the above. Generally, the hydraulic conductivity k of sand is larger than clay one and the distribution coefficient K_d of sand is smaller than clay.

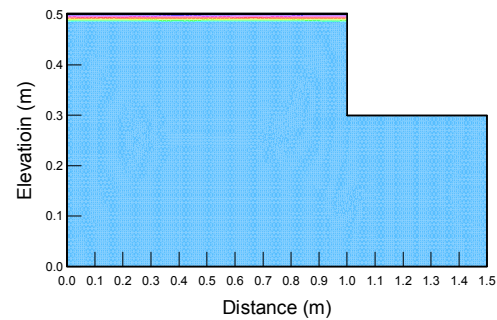


(a) 1 year progress

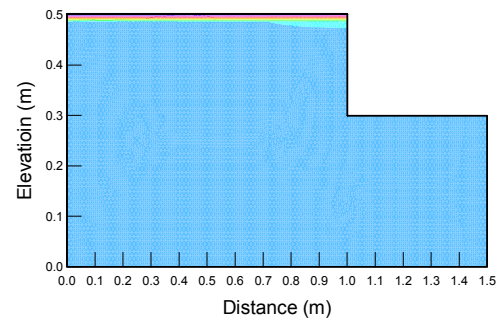


(b) 5 years progress

Fig. 5 The transition of Cs-137 in the sand layer



(a) 1 year progress



(b) 5 years progress

Fig. 6 The transition of Cs-137 in the clay layer

6.2 Results of the Analysis

Figures 5 and 6 show the transition of Cs-137 concentration in the sand and clay layers. In the sand layer, there is a large infiltration around the top of slope so that Cs-137 reaches by the foot of slope, placed depth 20cm, at five years progress. Consequently the current decontamination, digging and removing the ground layer about 5cm, has no effective in this case at 5 years progress. On the other hand, there is, on the whole, little infiltration in the clay layer, Cs-137 stays by outer layer 5cm. Therefore the current decontamination has effective in clay layer.

From the results, the differences of the infiltration behavior of radioactive substances occurs by the soils property. The decontamination should be selected by soils of each point.

7. CONCLUSIONS

In this study, a term that considered radioactive half-life embedded to advection-dispersion equation in the order to evaluate infiltration behaviors of radioactive substances in the soils. This analysis method was confirmed the validity for reproducing on the spot by comparing analysis results and measured results. Furthermore, Infiltration and advection-dispersion analysis was carried out in two cases, sand layer and clay layer. Infiltration and advection-dispersion analysis considered radioactive half-life can be applied to evaluate the current decontamination with infiltration and advection-dispersion analysis considered radioactive half-life. As a result, the decontamination should be selected by soils. Henceforth, it is necessary to evaluate the current decontamination from the analysis results that is almost the true condition, for example, implementing the infiltration and advection-dispersion analysis considered uncertainties and setting the analysis section modeled the real topography. Finally, Structure of final disposal site for decontaminated grounds should be evaluated by infiltration and dispersion-diffusion analysis which is considered radioactive half-life.

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