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RESEARCH PAPER

Transport Parameters of ^{99}Tc, ^{137}Cs, ^{90}Sr, and $^{239+240}$Pu for Soils in Korea

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Abstract : *To characterize quantitatively the transport of ^{99}Tc and the global fallout (^{137}Cs, ^{90}Sr, and $^{239+240}$Pu) for soils in Korea, the transport parameters of a convective-dispersion model, apparent migration velocity, and apparent dispersion coefficient were estimated from the vertical depth profiles of the radionuclides in soils. The vertical profiles of ^{99}Tc were measured from a pot experiment for paddy soil that had been sampled from a rice-field around the Gyeongju radioactive waste repository in Korea, and the vertical depth distributions of the global fallout ^{137}Cs, ^{90}Sr, and $^{239+240}$Pu were measured from the soil samples that were taken from local areas in Korea.*
The front edge of the ^{99}Tc profiles reached a depth of about 12 cm in 138 days, indicating a faster movement than the fallout radionuclides. A weak adsorption of ^{99}Tc on the soil particles by the formation of Tc(VII) and a high water infiltration velocity seemed to have controlled the migration of ^{99}Tc. The apparent migration velocity and dispersion coefficient of ^{99}Tc for the disturbed paddy soil were 2.88 cm/y and 6.3 cm^2/y, respectively. The majority of the global fallout ^{137}Cs, ^{90}Sr, and $^{239+240}$Pu were found in the top 20 cm of the soils even after a transport of about 30 years. The transport parameters for the global fallout radionuclides were 0.01-0.1cm/y (^{137}Cs), 0.09-0.13cm/y (^{90}Sr), and 0.09-0.18cm/y ($^{239+240}$Pu) for the apparent migration velocity: 0.21-1.09 cm^2/y (^{137}Cs), 0.12-0.7cm^2/y (^{90}Sr), and 0.09-0.36cm^2/y ($^{239+240}$Pu) for the apparent dispersion coefficient.

Keywords: *Transport in soil, Convective-dispersion model, Migration velocity, Dispersion coefficient*

1. INTRODUCTION

Owing to long half-lives and high radiotoxicity, the radionuclides, ^{137}Cs, ^{90}Sr, and $^{239+240}$Pu, persist in the environment for a long time after they have been released into the environment, and they can cause radiological damage to humans and biota through various transport pathways in the environment. ^{99}Tc is also known to be one of the important radionuclides that control the total radiation dose in a radiological safety assessment for the radioactive waste repository owing to their high mobility in soil and long half-lives. Thus, the transport parameters of such radionuclides in soil media is useful for appraising the soil contamination of radionuclides or the safety of surface ground disposal.

There are numerous studies on the radionuclide transport in soil, which have been performed by manifold approaches such as laboratory experiments, field observations, and modeling works. Ivanov et al. [1] investigated the distribution of ^{137}Cs and ^{90}Sr from the Chernobyl fallout in Ukrainian, Belarusian, and Russian soils and estimated the vertical transport parameters using a convective-dispersion model. To predict the transport of radionuclides in soil, Kirchner [2] proposed a stochastic model based on the probability density functions of solute displacement and applied the model to the depth profiles of ^{90}Sr and ^{137}Cs fallout measured in Orthic Podsol soil. Bossew and Kirchner [3] discussed some properties of the convective- dispersion model to predict the vertical transport of radionuclides in soil and statistically evaluated the transport parameters obtained for 528 measured radionuclides soil profiles in Austria. In particular, Shinonaga et al. [4] investigated the vertical profiles of ^{60}Co, ^{137}Cs, and ^{226}Ra in disturbed agricultural soils and calculated their transport parameters by means of a convective-dispersive model. McClellan et al. [5] studied the vertical transport of ^{228}Ac in a contaminated site in New Mexico in the USA and tested the compartment diffusion model and the leach rate model to determine their capabilities in predicting radionuclide behavior in soil. More recently, Solovitch-Vella et al. [6] studied the transport behaviors of global fallout ^{90}Sr, $^{239+240}$Pu,

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and ^{241}Am in mineral and organic soils in France. In that study, they applied a compartment model to obtain quantitative information on the vertical transport of the radionuclides.

In Korea, a number of attempts have been made to measure the vertical distribution of the global fallout radionuclides in undisturbed natural soils. Lee et al. [7, 8] studied the distribution and characteristics of the global fallout ^{137}Cs and $^{239+240}$Pu in undisturbed soils that were sampled from several local areas. Subsequently, some other studies were carried out to investigate the influence of the soil properties on the vertical distribution of the global fallout ^{137}Cs, ^{90}Sr, and $^{239+240}$Pu [9-13]. However, there are no transport parameters of radionuclides available for the soils in Korea. Our study estimated the transport parameters of the convective- dispersion model, apparent migration velocity and apparent dispersion coefficient to characterize quantitatively the transport of ^{99}Tc in a paddy soil and of the global fallout radionuclides (^{137}Cs, ^{90}Sr and $^{239+240}$Pu) in natural soils in Korea.

2. MODEL

The convective-dispersion model has been used to describe the transport behavior in saturated soils of chemical tracer or radionuclides [1, 3, 4, 14, 15]. The model is basically derived using the conservation law of mass.

$$\frac{\partial C}{\partial t} + \frac{\rho}{\phi}\frac{\partial q}{\partial t} = D\frac{\partial^2 C}{\partial z^2} - v_w\frac{\partial C}{\partial z} - \lambda(C+\frac{\rho}{\phi}q) \qquad (1)$$

where C (Bq/cm^3) is the flux-averaged concentration of radionuclides in groundwater; q (Bq/g) is the adsorbed concentration of radionuclides in soil; ρ (g/cm^3) is the bulk density of the soil; ϕ is the soil porosity; D (cm^2/y) is the effective axial dispersion coefficient; v_w (cm/y) is the interstitial water velocity; λ (y^{-1}) is the decay constant of a radionuclide.

The local equilibrium is often assumed between water and soil since the adsorption rate is generally faster than the moving velocity of the water in the soil. With the assumption of a linear equilibrium relationship ($q=K_dC$), Eq (1) is transformed to

$$\frac{\partial C}{\partial t} = D_\alpha\frac{\partial^2 C}{\partial z^2} - v_\alpha\frac{\partial C}{\partial z} - \lambda C \qquad (2)$$

where D (cm^2/y) is the apparent dispersion coefficient (= D/R_d); v_a (cm/y) is the apparent migration velocity (= v_w/R_d); is the retardation factor defined by, $R_d = 1+\frac{\rho}{\phi}K_d$ and K_d (cm^3/g) is the equilibrium distribution coefficient. Eq (2) is a well-known convective-dispersion model.

If $C_t(t,z)$ (Bq/cm^3) is the total volumetric concentration of a radionuclide in water and soil at a given time and soil depth, it is then related to the flux-averaged concentration C, as [3, 14]

$$C_t = C + \frac{\rho}{\phi}q = R_d C \qquad (3)$$

By combining Eqs. (2) and (3), the convective-dispersion equation for C_t is derived.

$$\frac{\partial C_t}{\partial t} = D_\alpha\frac{\partial^2 C_t}{\partial z^2} - v_\alpha\frac{\partial C_t}{\partial z} - \lambda C_t \qquad (4)$$

It can be seen that Eq. (2) is mathematically identical to Eq. (4) except that is replaced with C_t. With the following initial and boundary conditions, the solution of Eq. (4) is given by Eq. (5) [1-3].

IC : Ct = 0

BC : $v_a C_t(0^+, t) = (C_d\delta(t), C_t(\infty, t) \to 0$

$$\frac{C_t(z,t)}{C_d} = \frac{e^{-\lambda t}}{2}\left[-\frac{v_\alpha}{D_\alpha}\exp\left(\frac{v_\alpha z}{D_\alpha}\right)erfc\left(\frac{z+v_\alpha t}{2\sqrt{D_\alpha t}}\right)+\frac{2}{\sqrt{\pi D_\alpha t}}\exp\left(\frac{(z-v_\alpha t)^2}{4D_\alpha t}\right)\right] \qquad (5)$$

where C_d (Bq/cm^2) is the initially deposited amount of radionuclide on solid surface, and $\delta(t)$ (y^{-1}) is the Dirac delta function to simulate a pulse-like deposition of a radionuclide in soil as the result of an accidental release.

It is noted that the model parameters v_a and D_a in Eq. (5) incorporate internally all of the effects of the physicochemical properties of soil such as the porosity, organic matter content, and clay content, as well as climatic conditions such as the rainfall rate. The transport parameters were obtained from an optimization technique by applying Marquardt's algorithm [16].

3. MATERIAL AND METHODS

3.1 Transport of ^{99}Tc in Paddy Soil

Paddy soil (the main texture is silt loam, the pH is 5.1, and the organic matter content is 4.2%) was sampled from rice-fields around the Gyeongju radioactive waste repository in Korea. To use the soil in the transport experiment, it was dried, sieved to remove large gravel stones (> 2 mm), and homogenized.

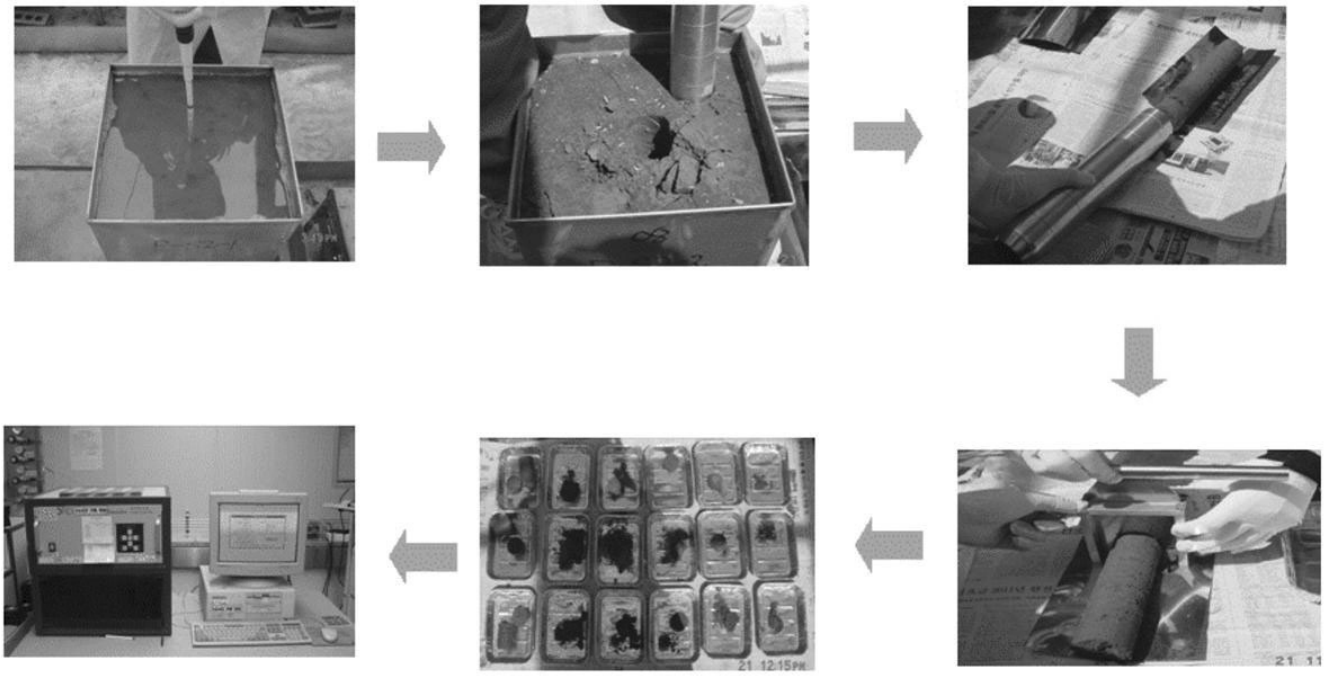

Fig. 1. Experimental steps for ^{99}Tc transport in a paddy soil.

Fig. 1 shows the experimental steps for ^{99}Tc transport in the paddy soil. Pretreated soil of about 31 kg was put in a stainless steel pot (dimensions: 30 cm × 30 cm × 40cm).

Prior to the transport, a sufficient amount of water was added to the pot until the soil was saturated in water. At the start of the transport, 5.3 kBq^{99}Tc/cm^2 was applied to the flooding surface water uniformly using a micropipette. During the transport, water was periodically added to the pot in an amount equal to the total amount of annual precipitation and irrigation water supplied in the paddy soil for rice cultivation in Korea. The amount of the water supply corresponds to a water interstitial velocity (v_w) of 0.55 cm/d [17]. One hundred thirty-eight days after the start of the transport, three core samples (5 cm in diameter) in the diagonal direction of the pot were taken up to a depth of 25 cm, and subsequently the samples were sectioned in at appropriate intervals (0-1, 1-3, 3-6, 6-10, 10-15, 15-25 cm) along the longitudinal axis to obtain the depth profiles of ^{99}Tc. The radioactivity of ^{99}Tc in each slice was analyzed by the total beta counter after being dried and ground. In the analysis of the radioactivity, the background activity of ^{99}Tc in the soil was ignored because it was so small compared to the total activity used in the transport experiment. The average value of the three diagonal core samples for each slice was taken as the measured activity at each depth.

3.2 Vertical depth profiles of the global fallout ^{137}Cs, ^{90}Sr and $^{239+240}$Pu

The measured vertical profiles (20 for ^{137}Cs, 6 for ^{90}Sr, and 6 for $^{239+240}$Pu) for global fallout radionuclides were taken from previous studies [7, 9-13], and they were fitted to the convective-dispersion model to estimate the transport parameters. Three adjusting model parameters (v_a, D_a, and C_d) were considered since the initially deposited amount of global fallout radionuclides into the soil (C_d) had been unknown. For model predictions, the transport time of the radionuclides should be previously known. Lee et al. [8] reported that the global fallout radionuclides in Korea mainly originated from a nuclear weapon test in the 1960s. Thus, the transport time of radionuclides for model predictions was assumed to be the interval between the sampling year of the soil and 1965 (the mid-year of the 1960s). For example, if a soil sample was taken in 2000, the migration time would then be 35 years.

4. RESULTS AND DISCUSSION

4.1 Transport of ^{99}Tc

Fig. 2 shows the measured profile of ^{99}Tc in the paddy

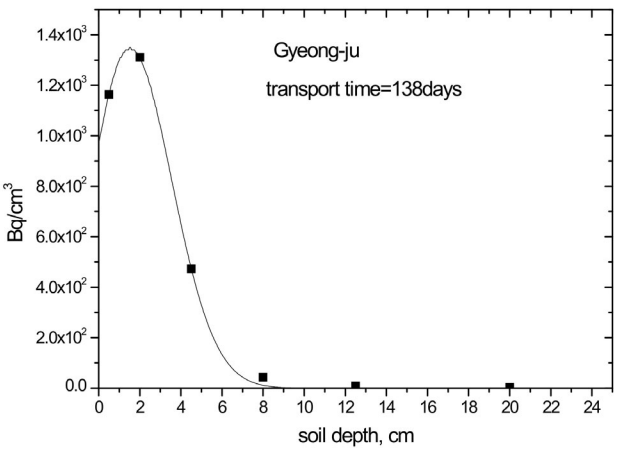

Fig. 2. Transport of ^{99}Tc in saturated paddy soil, Gyeongju, Korea.

soil at 138 days after the start of the transport and its comparison with the model prediction. The front edge of the ^{99}Tc profiles reached a depth of about 12 cm in 138 days. The fast movement of technetium in the paddy soil was quite different from the transport characteristics of fallout ^{99}Tc in the undisturbed natural soil. Tagami and Uchida [18] observed a very slow movement of the fallout ^{99}Tc in the undisturbed soils in Japan. They suggested that the slow movement of ^{99}Tc in the natural undisturbed soil would be attributed to the local reducing condition of the soil. It is well known that technetium changes into an insoluble low oxidation state and is immobilized in soil under reducing conditions [19]. On the other hand, when technetium is in an oxidizing condition, very soluble TcO_4^- is formed in water, and it migrates easily through soil. Not only soil bacteria but also soil organic matter is known to play a role in the formation of immobile Tc in soil [20-23]. In addition, a very high water interstitial velocity that was used in the present transport experiment, is thought to be responsible for the fast movement of the technetium in the paddy soil.

The best model prediction for the experimental profile of technetium in the paddy soil was obtained with the apparent migration velocity of 2.88 cm/y and the dispersion coefficient of 6.3 cm^2/y.

From the value of the apparent migration velocity (= v_w/R_d), the equilibrium distribution coefficient (K_d) of technetium for the paddy soil was calculated. The estimated K_d was 26.3 cm^3/g when the typical properties for the paddy soil in Korea ($\phi = 0.4$, ρ=1.04 g/cm^3, and v_w = 0.55 cm/d) were used. According to an IAEA [24] report, the Kd of technetium ranged from 0 to 100 cm^3/g under oxidizing conditions. Considering that the present Kd value of 26.3 cm^3/g exists in that range, local oxidizing conditions were likely to be maintained in the paddy soil during the present transport of technetium and consequently led to the facilitating migration of technetium as a result of the formation of soluble Tc(VII) in the paddy soil, in addition to the effect of a high water infiltration rate.

4.2 Transport of Global Fallout ^{137}Cs, ^{90}Sr, and $^{239+240}Pu$

Fig. 3 through 5 show some examples of the measured vertical profiles of the global fallout ^{137}Cs, ^{90}Sr, and $^{239+240}Pu$ for soils in Korea, and their comparison with the model predictions. The radionuclides seem to have moved to a deeper soil very slowly following the deposition onto the soil surface. The majority of radionuclides were observed in the top 20 cm of the soils, even after the transport of about 30 years. The transport parameters for the radionuclides that were obtained by fitting the convective-dispersion model are summarized in Table 1, together with their average value and standard deviation of the soils. Of the adjusting parameters, C_d is the initial deposited amount reduced to the sampling year of each soil, which is approximated to the total inventory of the soil sample at the sampling time if there was no major loss of radionuclide during transport. Most of the calculated C_d is in agreement with the measured total inventory (C_m) within a few percent of deviation, except for two soils, Gangleung and Seongsan, which had 37% and 15% deviation, respectively. The range of apparent migration velocity (v_a) was 0.01-0.1 cm/y for ^{137}Cs, 0.09-0.13 cm/y for ^{90}Sr, and 0.09-0.18 cm/y for $^{239+240}Pu$; and

Table 1. Transport parameters of global fallout ^{137}Cs, ^{90}Sr, and $^{239+240}Pu$ for soils in Korea

Location of soil sample	Soil sampling year	V_a(cm/y)	Da(cm^2/y)	C_d (*C_m) (Bq/cm^2)
^{137}Cs				
Boeun	1994	0.056	0.21	0.25 (0.25)
Ewang	1994	0.049	0.12	0.38 (0.32)
Daejeon	1994	0.065	0.41	0.08 (0.07)
Gangleung	1944	0.024	0.33	0.35 (0.22)
Goseong	2003	0.059	0.71	1.52 (1.48)
Jeju	2003	0.110	0.51	1.55 (1.44)
Uljin	2003	0.042	0.41	0.20 (0.29)
Jeongseon	1994	0.033	0.30	0.27 (0.26)
Seogipo	1996	0.060	1.09	0.91 (0.95)
Seongsan	1996	0.013	0.52	0.88 (0.75)
Ave. ± SD(%)		0.051 ± 52%	0.46 ± 60%	
^{90}Sr				
Ewang	1994	0.09	0.12	0.18 (0.17)
Seogipo	1996	0.13	0.70	0.38 (0.41)
Gongju	1994	0.11	0.22	0.20 (0.20)
Ave. ± SD(%)		0.071 ± 72%	0.35 ± 89%	
$^{239+240}Pu$				
Ewang	1994	0.09	0.09	0.01(0.01)
Seogipo	1996	0.18	0.30	0.024 (0.026)
Seongsan	1996	0.09	0.36	0.024 (0.022)
Ave. ± SD(%)		0.12 ± 43%	0.25 ± 56%	

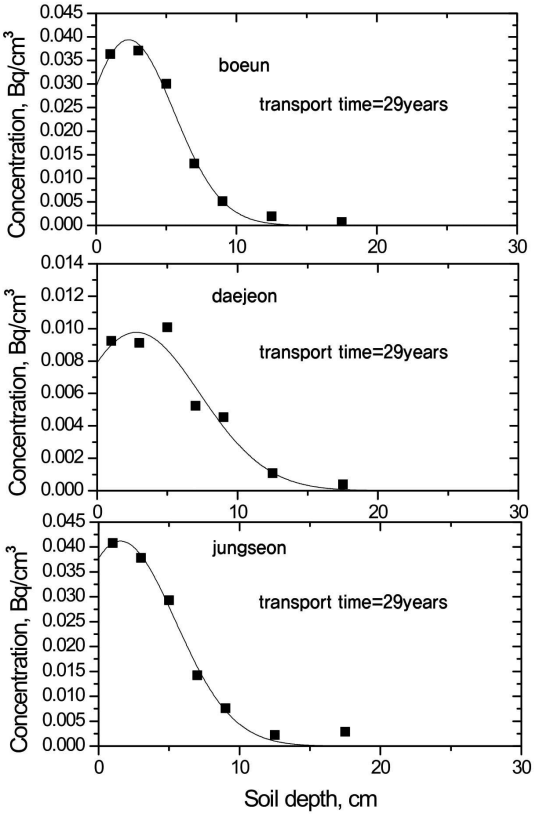

Fig. 3. Measured soil depth profile of ^{137}Cs and a comparison with the model predictions.

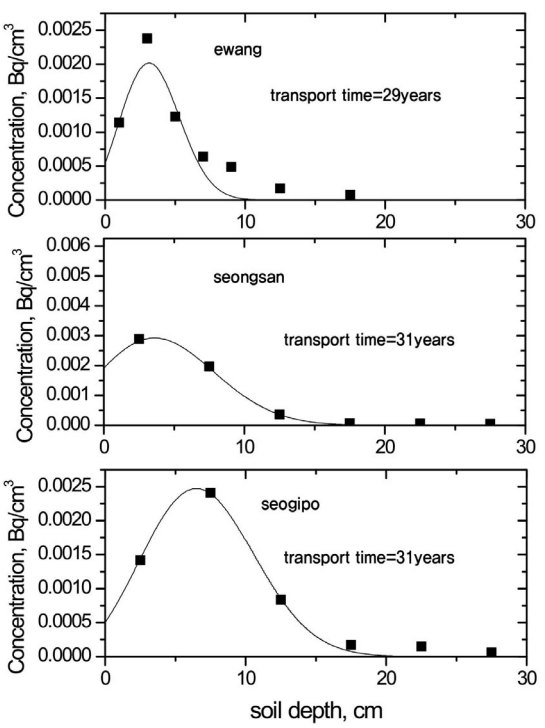

Fig. 4. Measured soil depth profile of $^{239+240}$Pu and a comparison with the model predictions.

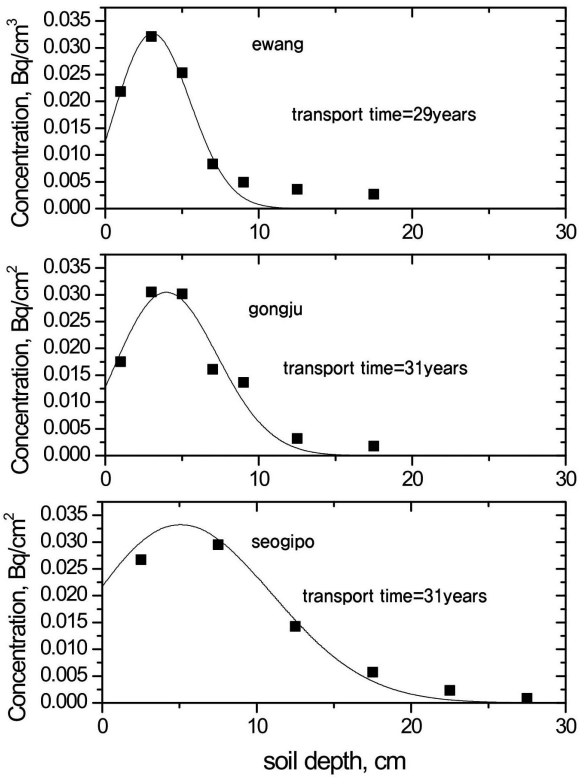

Fig. 5. Measured soil depth profile of ^{90}Sr and a comparison with the model predictions.

the range of the apparent dispersion coefficients (D_a) was 0.21-1.09 cm^2/y for ^{137}Cs, 0.12-0.7 cm^2/y for ^{90}Sr, and 0.09-0.36 cm^2/y for $^{239+240}$Pu, respectively. Our results partly overlap with the values of transport parameters obtained by other researchers. For example, Bossew and Kirchner [3] reported a range of the migration velocity of 0.1-0.5 cm/y and a dispersion coefficient of 0.05-0.5 cm^2/y for ^{137}Cs for soils in Austria. Shinonaga et al. [4] found a ^{137}Cs migration velocity of 0.0-3.0 cm/y for an agricultural disturbed soil. Solovitch-Vella et al. [6] reported a range of transport rates of 0.12-4.33 (^{90}Sr) and 0.14-3.5 cm/y ($^{239+240}$Pu) for mineral and organic soils in France. Ivanov et al. [1] reported the transport parameters of the Chernobyl fallout ^{137}Cs and ^{90}Sr for Ukrainian, Belarusian, and Russian soils: a migration velocity of 0.07-0.89 cm/y and a dispersion coefficient of 0.06-0.69 cm^2/y for ^{137}Cs, and a migration velocity of 0.06-0.92 cm/y and a dispersion coefficient of 0.14-1.73 cm^2/y for ^{90}Sr.

To investigate the difference in soil on the mobility of a radionuclide, the migration velocity of radionuclides was compared between two different soils, Ewang and Seogipo soil. The Ewang soil (pH 4.9, bulk density 1.2 g/cm^3, organic matter 11.3%, and clay 17.3%) and Seogipo soil (pH 6.1, bulk density 0.7 g/cm^3, organic matter 35.4%, and clay 21.2%) represent a typical granitic soil and volcanic soil in Korea, respectively. It is

interesting that all the ^{137}Cs, ^{90}Sr, and $^{239+240}$Pu had higher migration velocities in the Seogipo soil, in spite of the fact that both organic and clay content are higher in such soil. The organic matter can exhibit two opposite aspects for radionuclide migration in soil. The organic matter of a colloidal form in pore water can accelerate the migration of a radionuclide through the water flow, and at the same time the organic matter can retard the migration of a radionuclide when it is immobilized in soil. A study on the effect of organic matter content on radionuclide migration in Korean soils [8] showed a positive correlation between the concentration of ^{137}Cs and $^{239+240}$Pu in soil and organic matter, but this result cannot directly support the idea that a higher organic matter content would give rise to a lower migration rate because the colloidal effect of the organic matter was not simultaneously evaluated. Meanwhile, it is well known that a high clay content can restrict the mobility of ^{137}Cs in soil. However, in our study, the migration rate of ^{137}Cs was much higher in the Seogipo soil characterized by a higher clay content. This result does not indicate a contradiction to reality, but it simply suggests that other factors, rather than the clay content, would play a more important role in the difference in the migration velocity between two soils, since the migration velocity incorporates the combined effect of all variables involved in the radionuclide migration through soil. Another possible explanation of the difference in the transport between the soils is related to precipitation. The annual rainfall rate in the Seogipo area, 400 to 1900 mm for the past 30 years, is known to be larger than that in other regions of Korea, including the Ewang region. The higher rainfall rate in the Seogipo region is very likely to be the major reason for the higher migration velocity in the Seogipo soil. A high porosity of the volcanic soil could also be a main contributor to the higher migration velocity of radionuclides in the Seogipo soil.

In the meantime, in both the Ewang and Seogipo soils, the migration velocity of ^{90}Sr and $^{239+240}$Pu was higher by a factor of 2 or 3 than that of ^{137}Cs, which indicates a lower mobility of ^{137}Cs in those soils. That was possibly due to the well-known strong adsorption of ^{137}Cs in clay particles in soil. In the Ewang soil, both the migration velocity and dispersion coefficient were very similar for ^{90}Sr and $^{239+240}$Pu, whereas in the Seogipo soil, $^{239+240}$Pu exhibited a higher migration velocity and lower dispersion coefficient than ^{90}Sr. Although the colloidal formation owing to high organic matter in the Seogipo soil could be an explanation for the higher velocity of plutonium in the soil, it should remain only a suggestion until the effect can be clarified through future studies.

5. CONCLUSION

We fitted a convective-dispersion model to the vertical profiles of ^{99}Tc measured in saturated paddy soil and determined the vertical distributions of the global fallout ^{137}Cs, ^{90}Sr, and $^{239+240}$Pu in undisturbed natural soils to characterize quantitatively the radionuclide transport for soils in Korea

The apparent migration velocity of ^{99}Tc indicated a fast movement of radionuclides in the studied paddy soil. This seems to have resulted from a weak adsorption of ^{99}Tc on the soil particles, possibly owing to the existence of oxidizing conditions and a high infiltration velocity of water. The transport parameters of global fallout ^{137}Cs, ^{90}Sr, and $^{239+240}$Pu indicated a very slow movement in the undisturbed soils in Korea, and these results partly overlap with the values of the transport parameters obtained for different soils found by other researchers. The global fallout radionuclides exhibited a higher transport rate in volcanic soil compared with that in granitic soil. A higher rainfall rate and the porosity of the volcanic soil were very likely to be major reasons for the fast movement of radionuclides in volcanic soil. However, further studies are recommended to clarify the effect of variables involved in radionuclide transport in soil.

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