RETARDATION FACTOR OF RADIOACTIVE STRONTIUM IN A SANDY SOIL LAYER

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ABSTRACT

The retardation factor of ^{85}Sr for a sandy soil layer was determined to be a range from 1000 to 3700 by fitting the concentration distribution of ^{85}Sr calculated by the computer code of MIGSTEM-FIT to that observed in soil columns utilized in STEM (Environmental Simulation Test Program). Confidence intervals for the retardation factor were estimated based on statistical results. The distribution of retardation factors of ^{85}Sr for the soil layer is considered as a normal distribution. The minimum values of the confidence interval for the retardation factor are 8 to 26% smaller than the mean value. Thus, the rate of migration of ^{85}Sr could be underestimated by 8 to 26% if the mean value of the retardation factor was used.

INTRODUCTION

It is important to predict the migration of radionuclides in a shallow land system in order to estimate the impact of LLW shallow land disposal. The migration of radionuclides can be predicted by using parameters in the hydrodynamic transport equation for the element in question(1-3). A retardation factor is one of the important parameters for predicting the migration of radionuclides in groundwater through a soil layer. Therefore, the retardation factors of radionuclides must be measured to provide this input parameter for the prediction.

A retardation factor is usually determined in the laboratory using batch method based on the relationship between a retardation factor and a distribution coefficient(2,3). Retardation factors have also been determined by column method using collected soil layers(4). Bachhuber, et al. have compared retardation factors measured by a batch method and a column method using an undisturbed soil layer with in-situ retardation factors determined from field measurements(5). Their results showed that the retardation factors based on the field data were in accordance with that by the column method, but different from that by the batch method. Since the retardation factor based on the field data corresponds to the true migration rates of the radionuclides, the most accurate retardation factors must be measured in-situ or at nearly the same conditions as exist in-situ.

The Japan Atomic Energy Research Institute (JAERI) has conducted an "Environmental Simulation Test Program" named STEM, which examines the migration of radionuclides in large column of undisturbed soil. Therefore, a retardation factor closely matching in-situ conditions can be measured directly.

The migration of ^{60}Co, ^{85}Sr and ^{137}Cs has been carried out at the STEM facility using the sandy soil layer sampled. Of these radionuclides, the migration of Sr is predicted by using retardation factor(6). The migration mechanism of the ^{60}Co and ^{137}Cs is not expressed by the simple equilibrium ion exchanges reaction(7). Thus, the retardation factor of radioactive strontium for the sandy soil layer was determined in STEM.

METHOD

Migration model

The migration of radionuclides in a porous medium is a function of both the flow characteristics of the medium and the physical-chemical state of the radionuclides and the medium. Under the assumption that the porous medium is isotropic and homogeneous, the migration equation is expressed in the following form(8):

$$\frac{\partial}{\partial t}\left(C+\frac{\rho}{\theta}Q\right)=\nabla\cdot\boldsymbol{D}(\nabla C)-\nabla\cdot(\boldsymbol{V}C) \qquad (1)$$

where C : concentration of radionuclide in liquid phase,
Q : concentration of radionuclide in solid matrix,
D : dispersion coefficient,
V : water velocity,
ρ : bulk density of porous medium,
θ : water content.

The migration of the radionuclide can be predicted by Eq. (1), using the interaction between the solid phase and the radionuclide dissolved in the liquid phase. The interaction is expressed by the use of an adsorption isotherm if the interaction is caused by adsorption processes, and if equilibrium conditions of the interaction are established instantaneously. Several forms of isotherms are described in the literature(9). The simplest and most widely applied isotherm is the linear adsorption isotherm,

$$Q=Kd\ C \qquad (2)$$

where Kd is a distribution coefficient. Substituting Eq. (2) into Eq. (1), the equation takes the form:

$$\frac{\partial C}{\partial t}=\frac{1}{K_f}[\nabla\cdot\boldsymbol{D}(\nabla C)-\nabla\cdot(\boldsymbol{V}C)] \qquad (3)$$

where K_f is a retardation factor described by the following equation,

$$K_f = 1 + \frac{\rho}{\theta} K_d \qquad (4)$$

EXPERIMENTAL

Outline of STEM

Three undisturbed soil layers were obtained in 30 cm diameter by 120 cm length of columns at different depths ranging from 50 to 170 cm (sample A), from 170 to 290 cm (sample B) and from 290 to 410 cm (sample C) at the JAERI site. The soil layers were installed in the experimental facility of STEM for conducting migration experiments. A schematic diagram of the migration experiment apparatus is shown in Fig. 1.

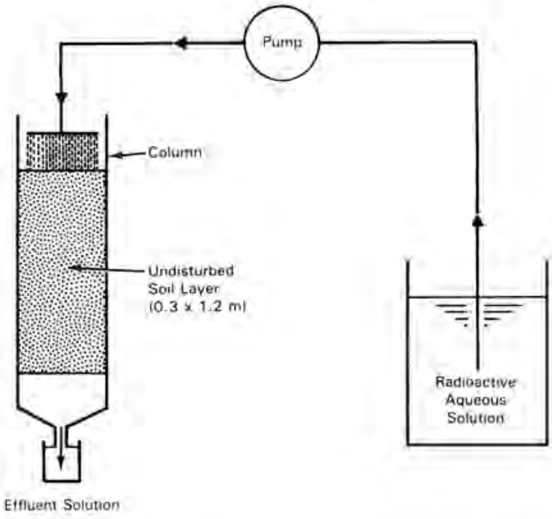


Fig. 1 Schematic diagram of column experimental apparatus in STEM.

Distilled water at pH 6.0 was spiked with ^{60}Co, ^{85}Sr and ^{137}Cs, and was passed into the soil column from the top at a constant flow rate of 1.5 ml/s. The use of distilled water was selected to simulate infiltrating moisture through the undisturbed soil layer. The concentration of each radionuclide was 3 uCi/L. The effluent solutions from the soil layer were collected at intervals of 2 hours. After the introduction of the radioactive aqueous solution, 15 to 21 separate soil cores of 5 cm in diameter were subsampled from each 30 cm diameter soil column to provide a statistical evaluation of the retardation factor. Soils were sampled at depth intervals of 2 cm in each soil core and the vertical one-dimensional concentration distributions of the radionuclides in each soil layer were. The activity of the radionuclides was measured by a gamma spectrum analyzer.

Determination of the retardation factor by fitting

When the water velocity and the dispersion coefficient are known, a retardation factor can be measured by fitting a concentration distribution in the soil layer or in the effluent solutions calculated by the mathematical migration model expressed by Eq. (3) to that obtained in a column experiment. The computer code (MIGSTEM-FIT) for determining a retardation factor by the fitting method has been developed in our laboratory(10). One must know values of the input parameters, such as water velocity and dispersion coefficient, as well as the initial and boundary conditions of the migration experiment, in order to obtain the concentration distribution of ^{85}Sr in the soil layer or the effluent distribution by using MIGSTEM-FIT.

The water velocities through the soil columns A, B and C were measured by the fast neutron transmission method(11) are presented in Table I. The dispersion coefficient is correlated to the dispersivity, Dm, by the following equation(12):

$$D = D_m V^{1.2} \ .$$

The dispersivities for the undisturbed soil layers were obtained by passing tritiated water through the column(12) and the resulting dispersivities are shown in Table I.

Table I

Parameters for determining retardation factor by MIGSTEM-FIT.

Sample	Water velocity (cm/s)	Dispersivity (cm)
A	2.0×10^{-3}	0.29
B	2.1×10^{-3}	0.23
C	2.8×10^{-3}	0.19

Since no radionuclide existed in the soil layer before the radioactive solution was introduced, the initial condition becomes:

$$C=0, \ t<0.$$

The radioactive solution continuously passed through the soil layer from the top. The boundary condition is

$$C_0=3.0 \ \mathrm{uCi/ml}, \ t>0.$$

At the bottom, the boundary condition is

$$dC/dz=0.$$

RESULT

Figure 2 presents the concentration distribution of ^{85}Sr observed in the soil layer of sample B. The concentrations of ^{85}Sr in the effluent solutions were less than 10^{-4} uCi/L. This indicates that most of the ^{85}Sr remained in the soil layer. The retardation factor of ^{85}Sr can be determined by fitting the observed concentration distribution of ^{85}Sr in the soil layer using the MIGSTEM-FIT program. The solid line in Fig. 2 indicates the concentration distribution of ^{85}Sr in the soil layer calculated by the MIGSTEM-FIT based on the retardation factor of 2034. The concentration distribution in the soil layer calculated is in good agreement with that observed. This shows that the retardation factor determined by the MIGSTEM-FIT corresponds to that of ^{85}Sr for the soil.

The frequency histograms for the retardation factors of ^{85}Sr determined by replicate measurement using the soil layers of samples A, B and C are shown in Fig. 3. The mean values and standard deviations of the retardation factors of ^{85}Sr for the different depths of the soil are presented in Table II. Figure 3 indicates that the retardation factors for the soil layer of samples A, B and C have values in the range from 1000 to 2800, from 1400 to 3400, and from 1000 to 3700, respectively. The distribution is considered as a normal distribution. These results reveal:

1. The migration of ^{85}Sr through the three soil layers cannot be predicted precisely by using one retardation factor.

2. The migration of ^{85}Sr through each of the three soil layers cannot be predicted precisely by using the mean value of the retardation factor.

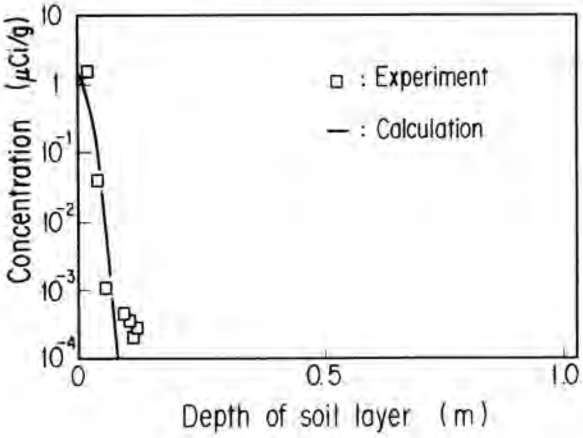
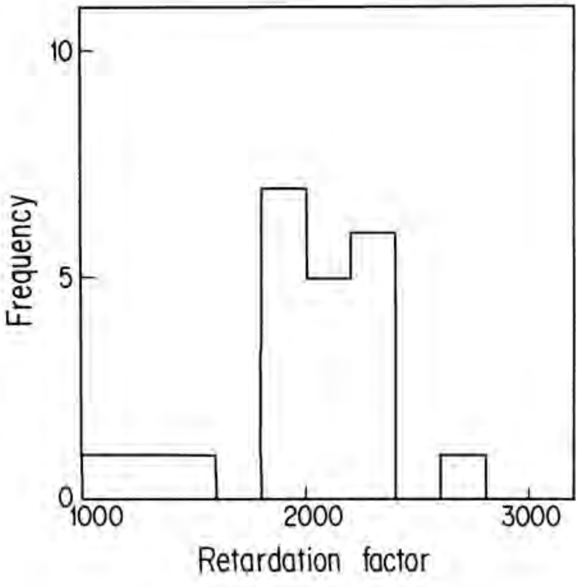
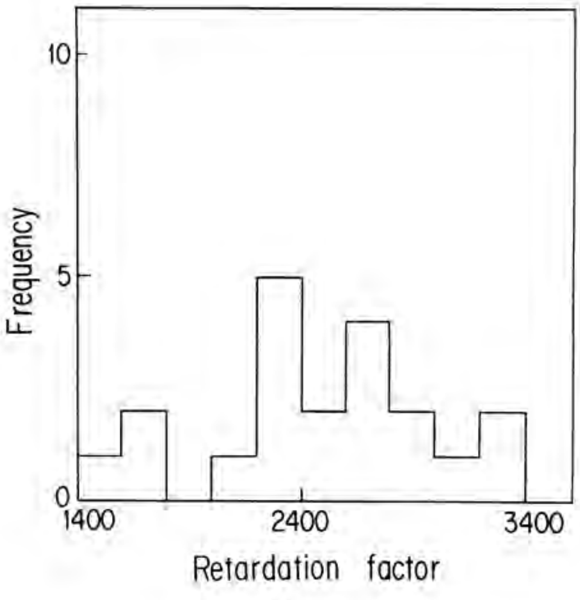


Fig. 2 Concentration distribution of ^{85}Sr in the soil layer observed in STEM and that calculated by MIGSTEM-FIT.

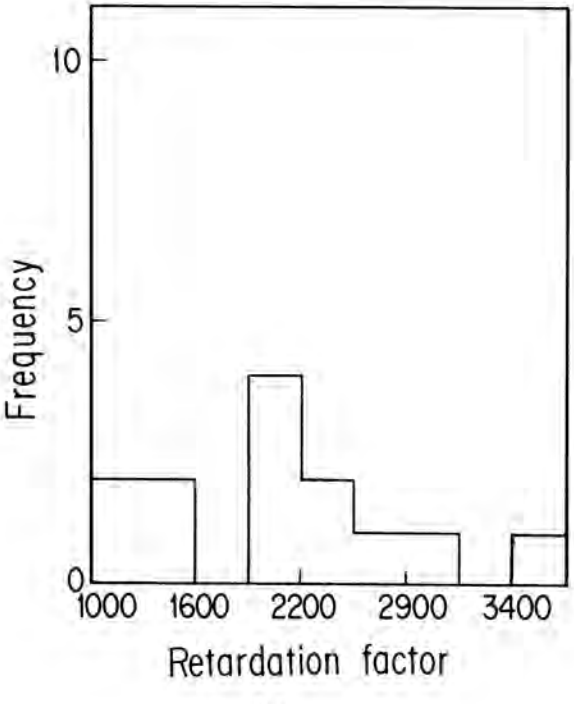
The retardation factor is a function of the distribution coefficient, water content and bulk density of the soil (Eq. (4)). Variations in three parameters for each soil layer of samples A, B and C were probably the reason for the observed differences in the retardation factors.



(a)



(b)



(c)

Fig. 3 Frequency histograms for retardation factor of ^{85}Sr measured for the soil layer of samples A: (a), B: (b) and C: (c).

Discussions

The 15 to 21 replicable measurement of the retardation factors for each soil layer resulted in a normal distribution. Therefore, we must estimate the true retardation factor of ^{85}Sr for the soil layer (population).

If a set of the retardation factors obtained by replicable experiments is expressed by a normal distribution function, the confidence interval for the mean of the population can be estimated by statistical technique. It is possible to determine the confidence interval for the retardation factor of ^{85}Sr obtained in the present study, because the retardation factor distributes normally.

Since the retardation factors were determined for the soil layer of samples A, B and C, the confidence intervals for the retardation factors should be estimated for each soil layer of the three samples independently. The method for determining the confidence interval for the retardation factor was presented in the literature(13).

Table II

Mean values and standard deviations of retardation factor of ^{85}Sr for the soil layers of sample A, B, and C.

Sample	Numbers of Rd measured	Mean value	Standard deviation
A	21	2012	364
B	21	2420	596
C	15	2438	1105

Table III

95 % confidence intervals for retardation factor of ^{85}Sr for the soil layers of sample A, B and C.

Sample	Confidence interval
A	1842 - 2183
B	2142 - 2698
C	1805 - 3071

The confidence intervals for the retardation factors of ^{85}Sr were estimated using the t-distribution, because the variance of the population was unknown. The 95 % confidence intervals are shown in Table III. The minimum values of the 95 % confidence intervals in Table III are important for the prediction of the migration of ^{85}Sr in the soil layer, because a smaller retardation factor gives a more conservative estimate of the rate of migration. The prediction of the migration of ^{85}Sr in the soil layer has a reliability of 95 %, if the 95 % confidence interval is used for the retardation factor. The minimum values in the confidence intervals of the retardation factors are smaller by 8.4, 11.5, and 26.0% than the mean values of the retardation factors of sample A, sample B and sample C, respectively. This shows that the prediction of the migration of ^{85}Sr in the soil layer may be underestimated. when we use the mean value of the retardation factor for the parameter. Therefore, it is prudent to measure the confidence interval for the retardation factor of radionuclides on the prediction of the migration of radionuclides.

CONCLUSIONS

The retardation factors of ^{85}Sr for a sandy soil layer were determined by fitting the concentration distribution of ^{85}Sr calculated by the computer code of MIGSTEM-FIT to that observed in STEM. The following conclusions were drawn:

1. The concentration distribution in the soil layer calculated by MIGSTEM-FIT was in good agreement with the observed distribution. This shows that the retardation factor determined by MIGSTEM-FIT can be used for estimating the migration of ^{85}Sr in the soil layer.

2. The retardation factors have values ranging from 1000 to 3700. This indicates that the migration of ^{85}Sr in the soil layer is not predicted precisely by using the mean value of the retardation factor.

3. The 95 % confidence interval for the retardation factors of ^{85}Sr for the soil layer were estimated using t-distribution. The minimum values of the confidence intervals in the retardation factors are smaller by 8.4, 11.5 and 26% than the mean values of the retardation factor of sample A, sample B, and sample C, respectively.

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