AN IN-SITU TEST INVESTIGATING THE MIGRATION OF RADIONUCLIDES ^{85}Sr, ^{134}Cs AND ^{60}Co IN UNSATURATED CHINESE LOESS

By Chunli Liu, Zhiming Wang, Yongxing Zhang*, Shanggeng Luo*,
Shunshen Li, Zede Guo and Ling Jiang
China Institute for Radiation Protection
PO Box 120 Taiyuan, Shanxi 030006, PR China

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ABSTRACT

As part of a co-operative research project between the China Institute for Radiation Protection (CIRP) and the Japan Atomic Energy Research Institute (JAERI) on Safety Assessment Method for Shallow Land Disposal of Low Level Radioactive Wastes, an in-situ test investigating the migration of radionuclides ^{60}Co, ^{85}Sr and ^{134}Cs in unsaturated Chinese loess under natural rainfall and artificial rainfall conditions was carried out. ^{3}H was used for obtaining the migration velocity of moisture water in the tested zone.

During the testing period, vertical soil core samples were taken 4 times a year in the experimental pits (A, B, D and E). In the first 2 months of applying artificial rainfall, soil water samples were sucked directly from pits D and E. When the 2 years in situ test was completed, 5 vertical core samples were taken in each of the 4 pits, respectively. Totally 47 core samples were obtained.

The core samples were cut into slices of 0.5 to 2 cm in thickness for laboratory analysis. When a core sample was taken, the hole left was filled with an end-closed iron tube to avoid the possible effect of the hole to the migration of radionuclides.

The concentration distribution of ^{3}H in artificial rainfall condition obtained with sucked water samples suggest that the hydraulic property of the backfilled part was different from the undisturbed part and from pit D to pit E, due to the backfilling. The ^{3}H concentration distribution obtained from soil core samples in the natural condition indicated the existence of a downward and upward movements of moisture water in rainy and dry seasons, respectively.

The result of the 2-year in-situ test indicates that the positions of peak concentration of ^{134}Cs and ^{60}Co in both the natural and the artificial rainfall conditions remained within 1 cm from their origins, due to the strong sorption of these two nuclides to the loess medium, while the position of the peak concentration of ^{85}Sr in the artificial rainfall condition moved down to a depth of about 12 to 18 cm. In natural rainfall condition, there was a triple peak concentration distribution for ^{85}Sr that might be caused by the downward and upward movements of moisture water in different seasons.

* China Institute of Atomic Energy

INTRODUCTION

Under an Agreement between the Governments of the People's Republic of China and Japan on Peaceful Use of Nuclear Energy, an Arrangement between the Bureau of Foreign Affairs of the Ministry of Nuclear Industry of the People's Republic of China and the Japan Atomic Energy Research Institute was made to conduct a Co-operative Research on Assessment Method of Radiological Safety for Shallow Land Disposal of Low Level Radioactive Waste. According to the arrangement, a Co-operative Research Project between the China Institute for Radiation Protection (CIRP) and the Japan Atomic Energy Research Institute (Tokai Establishment) on Safety Assessment Method for Shallow Land Disposal of Low Level Radioactive Wastes (LLW) was conducted. In the framework of the Co-operative Research Project, laboratory simulation tests, laboratory batch experiments and in-situ tests were performed investigating the migration of ^{85}Sr, $^{134/137}Cs$ and ^{60}Co in unsaturated Chinese loess. The main result of the project was described elsewhere[1]. This paper introduces the result of a 2-year in-situ test that was conducted at the CIPR's field test site in both natural and artificial rainfall conditions.

EXPERIMENTAL

The Field Test Site and the Experimental Pits

The field test site situates at a medium cut loess plateau with an elevation of 953 m, an average annual precipitation of 434 mm , an average annual evaporation of 326 mm , and an annual mean temperature of 9.3 ^0C [1].

The in-situ test was carried out in the field test site with experimental pits in both the natural rainfall and artificial rainfall conditions [2, 3]. Table 1 lists the dimensions of each experimental pit, the experimental conditions and the radionuclides introduced.

Table 1. Pit dimension, radionuclides and experimental condition [1].

Pit	Dimensions	Radionuclides	Exp. Condition
A	200×200×30 (Depth) cm^3	^3H,^{60}Co,^{85}Sr and ^{134}Cs	Natural rainfall
B	200×200×100 (Depth) cm^3	^3H,^{60}Co,^{85}Sr and ^{134}Cs	Natural rainfall
C	200×200×100 (Depth) cm^3	^{60}Co,^{85}Sr and ^{134}Cs	Natural rainfall
D	200×200×30 (depth) cm^3	^3H,^{60}Co,^{85}Sr and ^{134}Cs	Artificial rainfall
E	200×200×100 (depth) cm^3	^3H,^{60}Co,^{85}Sr and ^{134}Cs	Artificial rainfall
F	200×200×100 (depth) cm^3	^{60}Co,^{85}Sr and ^{134}Cs	Artificial rainfall

Instrumentation

In order to measure the meterological and hydrological parameters of the test site, instruments such as Electrical anaemmometer (E-L electrode), Siphon raingauge (Model SJ1), Thermograph (Model DWJ1), Thermograph (Model D1+J1), Soil thermometer, Tensiometer, neutron probe, etc were installed.

Tracer Layer

Solutions containing ^{60}Co, ^{85}Sr, ^{134}Cs (in the forms of $CoCl_2$, $SrCl_2$ and CsCl) and ^{3}H were used as radioactive solutions. The soil taken from the same pit was wind-dried and ground into sizes of 60 to 100 mesh. Put the ground soil into an electric stirrer and run the stirrer for a while to make an even mixing, then the radioactive solution with fine droplets was sprayed into the stirrer under a set pressure of 0.2 kg/cm^2 while the stirrer was running. In order to make a complete mixing of the radioactive solution with the soil, the stirrer continued to run for 30 minutes when the spraying was stopped. In order to know the exact amount of radioactivity in each mixture, 5 random samples were taken from each mixture at different points and analysed with gamma spectrometer. Table 2.lists the total amount of the 5 random samples taken from each mixture for pit A, B, D and E. As for pit C and F, 20 g of the ground soil was mixed with radioactive solution directly and dried to a weight wetness of about 15% with infrared heat and analysed with gamma spectrometer for total radioactivity [4]. The amount of ^{3}H introduced into each pit was calculated based on the volume and concentration of ^{3}H solution, because a considerable amount of ^{3}H would be evaporated out of the mixture during the tracer layer introduction process due to the dry weather condition at the test site. Table 3.lists the total activity of each radionuclide in each pit.

Table 2. Total amount of 5 random samples

Mixture	pit A	pit B	pit D	pit E
Total amount (g)	105	160	78.5	77.2

Table 3. Total amount of each radionuclide in each pit (Bq) [1].

	^{3}H	^{60}Co	^{85}Sr	^{134}Cs
Pit A	7.4E08	1.85E07	5.11E07	5.59E07
Pit B	7.4E08	1.92E07	4.85E07	6.25E07
Pit C		8.51E06	3.63E07	7.84E07
Pit D	7.4E08	1.70E06	4.63E07	8.73E07
Pit E	7.4E08	5.18E06	6.07E07	3.07E07
Pit F		4.44E06	7.40E07	3.22E07

Tracer Layer Introduction

Bottom-levelled pits (A, B, D and E) were dug and installed with tensiometers and water suckers on the bottom of each pit. Fix a square-steel-frame of 1.5×1.5 m on the bottom of each pit. Introduce the mixture into the frame and make it level. Scatter a thin quartz sand layer above the mixture and backfill the pit for several cm with the same soil dug out of the pit at each time. Install other tensiometers above the mixture and backfill the pits completely. For pit C and F, point tracer sources were introduced for direct gamma measurement and PVC tubes were installed for the direct detection of the gamma rays emitted [2]. Table 4 lists the dimensions of tracer layers and the density of the artificial rainfall.

Table 4. Tracer layer dimension and artificial rainfall density

Pit	Tracer layer	Rainfall density	
A	150 (cm) ×150 (cm) ×7(mm, thickness)	Natural Rainfall	
B	150 (cm) ×150 (cm) ×7(mm, thickness)	Natural Rainfall	
C	5.5 (cm, diameter) ×6(mm, thickness)	Natural Rainfall	
D	150 (cm) ×150 (cm) ×7(mm, thickness)	5mm/h,200×200 cm^2,3h/d	(741 days)
E	150 (cm) ×150 (cm) ×7(mm, thickness)	5mm/h,200×200 cm^2,3h/d	(741 days)
F	5.5 (cm, diameter) ×6(mm, thickness)	5mm/h,200×200 cm^2,3h/d	(750 days)

Sampling

In order to obtain the concentration distribution of these radionuclides in the soil during the testing period, vertical core samples were taken 4 times a year in pits A, B, D and E, respectively. During the first 2 months of applying artificial rainfall, soil water samples were sucked directly from pits D and E. The amount of soil water sucked was 2 ~ 20 ml/d. In some cases, no water was sucked. When the 2-year in-situ test was completed, 5 vertical core samples were taken in each of the 4 pits, respectively. Totally 47 core samples were obtained. Table 5 lists the parameters of some of the core samples. For pits C and F, direct detection methods for the distribution profiles were used, the result was introduced in [4].

Table 5. Parameters of some of the core samples

Pit.	Sample No	Test period (d)	Length of the Sample (cm)	Analysed nuclides
A	A-11	780	192	^{3}H,^{60}Co,^{85}Sr and^{134}Cs
A	A-12	780	175	^{3}H,^{60}Co,^{85}Sr and^{134}Cs
A	A-13	780	193	^{3}H,^{60}Co,^{85}Sr and^{134}Cs
A	A-14	780	198	^{3}H,^{60}Co,^{85}Sr and^{134}Cs
A	A-15	780	192	^{3}H,^{60}Co,^{85}Sr and^{134}Cs
B	B-11	779	191	^{3}H,^{60}Co,^{85}Sr and^{134}Cs
B	B-12	779	182	^{3}H,^{60}Co,^{85}Sr and^{134}Cs
B	B-13	779	197	^{3}H,^{60}Co,^{85}Sr and^{134}Cs
B	B-14	779	205	^{3}H,^{60}Co,^{85}Sr and^{134}Cs
B	B-15	779	185	^{3}H,^{60}Co,^{85}Sr and^{134}Cs
D	D-11	741	91	^{60}Co,^{85}Sr and^{134}Cs
D	D-12	741	105	^{60}Co,^{85}Sr and^{134}Cs
D	D-13	741	108	^{60}Co,^{85}Sr and^{134}Cs
D	D-14	741	110	^{60}Co,^{85}Sr and^{134}Cs
D	D-15	741	92	^{60}Co,^{85}Sr and^{134}Cs
E	E-11	741	184	^{60}Co,^{85}Sr and^{134}Cs
E	E-12	741	98	^{60}Co,^{85}Sr and^{134}Cs
E	E-13	741	203	^{60}Co,^{85}Sr and^{134}Cs
E	E-14	741	199	^{60}Co,^{85}Sr and^{134}Cs
E	E-15	741	185	^{60}Co,^{85}Sr and^{134}Cs

The core samples were cut into slices of 0.5 to 2 cm in thickness for laboratory analysis. When a core sample was taken, the hole left was filled with an end- closed iron tube to avoid the possible effect of the hole to the migration of radionuclides.

In pit D and E, "suckers" were used to obtain moisture water samples for estimating the moisture water movement velocity under artificial rainfall conditions. Table 6 lists the parameters of the sucked water samples.

Table 6. Parameters of the sucked water samples

Sampling pit & No	Sampling depth (cm)	Sampling period
D-1	60	July. 27~Sep. 5
D-2	110	Aug. 3 ~ Sep 10
D-3	150	Aug. 3 ~ Sep. 22
E-1	110	Aug. 7 ~ Sep. 22
E-2	150	Aug. 7 ~ Sep. 22

Sample Analysis

A gamma spectrometer system was used to analyse the radioactivity of ^{60}Co, ^{85}Sr and ^{134}Cs in the sliced soil samples, while a liquid scintillation counter was used to measure the radioactivity of ^{3}H in some sliced soil samples and in soil water samples. The detailed analysis methods were introduced in [5].

RESULTS AND DISCUSSION

Sucked ^{3}H Concentration Distributions in Pits D and E

Fig. 1 shows the concentration of ^{3}H in pits D and E as a function of the sucking time in the first two months of applying artificial rainfall. It indicates that the average water migration velocity in the backfilled part in pit D is different from that in pit E and the water migration velocity in the undisturbed part in pit D varies with depth. These suggest that the hydraulic property of the backfilled part might be different from the undisturbed part and different from pit D to pit E. That is, we did not have a satisfactory backfilling and we did not have a fixed water migration velocity at different depth while we had a fixed surface water infiltration flux in the in-situ test under artificial rainfall condition. The hydraulic property of the backfilled part is different from the undisturbed part was probably produced by backfilling techniques. The cause to the different average migration velocity of moisture water at different depth with a fixed surface infiltration flux is complex. A probable explanation is that because the surrounding of the pits is in natural condition, and the water content varies with depth, thus the amount of water horizontally dispersed varies with depth Table 7 lists the average water migration velocity calculated from Fig. 1 and Table 6. Table 8 lists the average migration velocity at different intervals calculated from Table7.

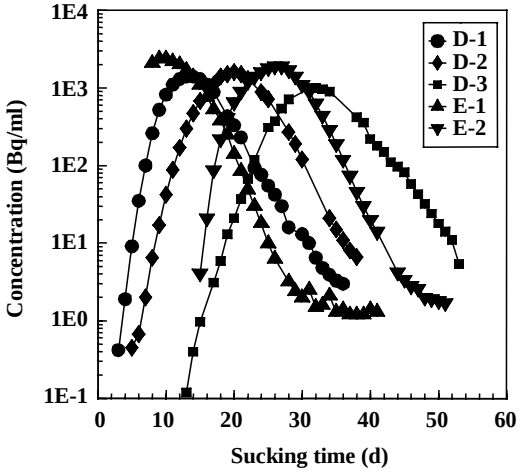

Fig1. The concentration of ^{3}H in pit D and E as a function of sucking time

Table 7. The average water migration velocity

Sucking point	Sucking depth (cm)	Peak arrival time (d)	Average velocity (cm/d)
D-1	60	23	2.6
D-2	110	30	3.7
D-3	150	42	3.6
E-1	110	19	5.8
E-2	150	36	4.2

Note: The artificial rainfall had been applied 20 days when the first sucking took place.

Table 8. Migration velocity at different intervals

Depth interval (cm)	Time interval (d)	Average Velocity (cm/d)
Surface/D1, 60	23	2.6 (30 cm backfill)
D1/D2, 50	7	7.1 (undisturbed)
D2/D3, 40	12	3.3 (undisturbed)
Surface/E1, 110	19	5.8 (100 cm backfill)
E1/E2, 40	17	2.4 (undisturbed)

Table 8 indicates that there is a big difference in the migration of water at intervals from 60 cm to 110 cm and from 110 cm to 150 cm in pit D.

^{3}H Concentration Distributions in Core Samples in Pits A and B

Fig. 2 and 3 show the ^{3}H concentration distributions in core samples obtained in pits A and B, respectively, when the 2 year in-situ test was completed. Fig. 2 indicates that in pit A most of the ^{3}H introduced moved down to depths of 30 to 60 cm (in the range of 60 to 90 cm from the surface), while Fig. 3 indicates that in pit B, part of the ^{3}H moved down to a depth of 40 cm, while part of them moved up 20 cm (in the range of 80 to 120 cm from the surface). Fig. 3 might suggest that at the field test site the downward movement of the moisture water at 100 cm from the surface was complex. In rainy seasons, there would be a downward movement while in dry seasons, there would be an upward movement. This downward and upward movement of moisture water may be one of the reasons inducing the triple peak concentration distribution of ^{85}Sr in pits A and B.

Concentration Distribution of ^{60}Co, ^{85}Sr and ^{134}Cs in Core Samples

Fig. 4 and 5 show two sets of the concentration distributions of ^{60}Co, ^{85}Sr and ^{134}Cs in core samples obtained from pits A and E, respectively. These figures indicate that in the artificial rainfall condition (Fig. 4), in more than 2 years, most of ^{60}Co and ^{134}Cs introduced remain within 1 cm from their origins, while ^{85}Sr moved down to a depth of about 12 to 18 cm.

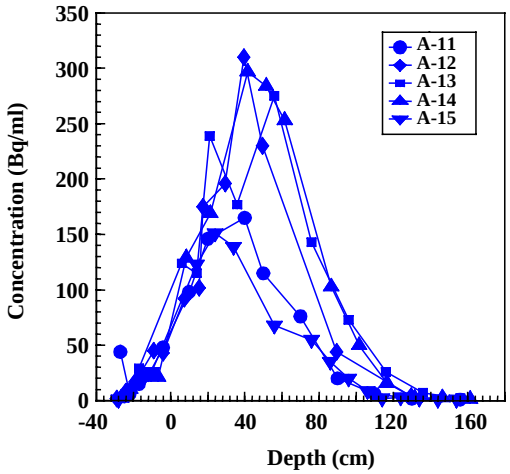

Fig2. The concentration of ^{3}H in pit A as a function of depth

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In the natural rainfall condition (Fig. 5), in more than 2 years, most of the ^{60}Co and ^{134}Cs remain within 1 cm from their origins like that in the artificial rainfall condition, while ^{85}Sr has three peaks, one peak moved down to a depth of 2 to 6 cm, one remained at its origin and one moved up. This triple peak phenomenon is probably caused by the downward and upward movements of moisture water in rainy and dry seasons during the test period.

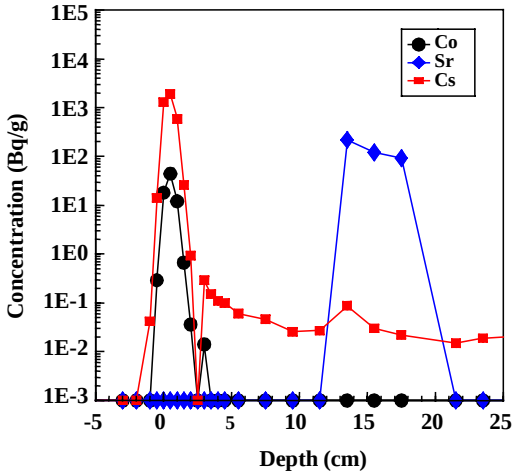

Fig4. The concentration of ^{60}Co, ^{85}Sr and ^{134}Cs as a function of depth in pit E

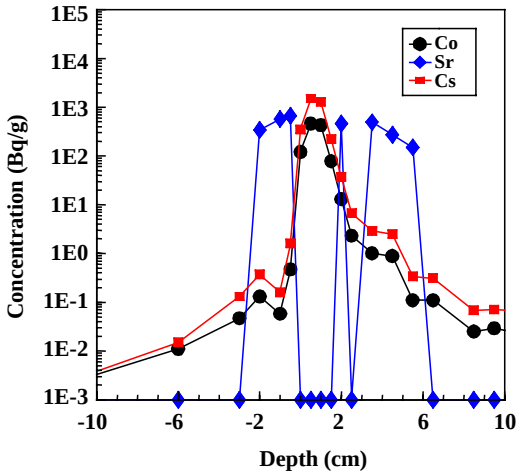

Fig5. The concentration of ^{60}Co, ^{85}Sr and ^{134}Cs as a function of depth in pit A

CONCLUSIONS

As part of a co-operative research project between CIRP and JAERI on Safety Assessment Method for Shallow Land Disposal of Low Level Radioactive Wastes, an in-situ test was carried out investigating the migration of radionuclides ^{60}Co, ^{85}Sr and ^{134}Cs in unsaturated Chinese loess. ^{3}H was used for obtaining the migration velocity of moisture water in the tested zone. The concentration distribution of ^{3}H in artificial rainfall condition obtained with sucked water samples suggest that the hydraulic property of the backfilled part might be different from the undisturbed part and from pit D to pit E, due to the backfilling. The ^{3}H concentration distribution obtained from soil core samples in the natural condition indicated the existence of a downward and upward movements of moisture water in rainy and dry seasons, respectively. This might be one of the reasons inducing the triple peak concentration distribution of ^{85}Sr in the natural condition. The result of the 2-year in-situ test indicates that the positions of peak concentration of ^{134}Cs and ^{60}Co in both the natural and the artificial rainfall conditions remained within 1 cm from their origins, due to the strong sorption of these two nuclides to the loess medium, while the position of the peak concentration of ^{85}Sr in the artificial rainfall condition moved down to a depth of about 12 to 18 cm. In natural rainfall condition, there was a triple peak concentration distribution for ^{85}Sr that might be caused by the downward and upward movements of moisture water in different seasons.

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