

Distribution of ^{90}Sr Activities in the Environmental Radiation Samples of Jeju Island, Korea

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ABSTRACT

Background: This work was to get information about ^{90}Sr contamination of the environment by using soil and moss from selected areas in Jeju Island, Korea.

Materials and Methods: The activities of ^{90}Sr in soil and moss samples were investigated at nine locations of Jeju island, Korea. The soil samples have been collected at 4 sites of Jeju island during June to August of 2013, analyzed for vertical distribution of ^{90}Sr activities. The moss samples have been collected at 5 sites of Jeju island during November of 2011 to June of 2012, and analyzed for radioactive ^{90}Sr .

Results and Discussion: The ^{90}Sr vertical concentrations in the investigated soil samples were 2.77 to 18.24 Bq·kg⁻¹ in eastern part, 1.69 to 18.27 Bq·kg⁻¹ in northern part, 3.76 to 13.46 Bq·kg⁻¹ in the western part and 1.09 to 8.70 Bq·kg⁻¹ in the southern part of the Mt. Halla in Jeju island, respectively. Activities of ^{90}Sr show the highest value at the surface soil and decrease with depth. The activity concentration measured was in the range of 79.6 to 363 Bq·kg⁻¹ -dry moss.

Conclusion: This material is expected to be basis reference for survey of environmental radioactivity in Jeju Island.

Keywords: Liquid scintillation counting, Radiostrontium, Soil, Moss

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Introduction

Some of the radionuclides that had been released to the atmosphere from the Fukushima Dai-ichi Nuclear Power Plant accident were transported to the Korean territory [1]. They included such radionuclides as ^{134}Cs , ^{137}Cs , ^{131}I , and ^{90}Sr . It was not known, however, whether ^{90}Sr had also reached to Korea or not, since ^{90}Sr and its daughter ^{90}Y are pure beta emitters and thus cannot be detected by gamma spectrometry. Conventional measurement of ^{90}Sr in environmental samples involves a lengthy chemical process. ^{90}Sr constitutes a long-term biological hazard because it accumulates in bone tissues, and has long physical and biological half-lives; 28.6 and 49.3 yr, respectively [2].

Nearly 99% of radioisotopes from atmosphere go to soil through precipitation. Absorbed by soil radioactive elements may move with soil solutions horizontally and vertically or stay in root region of plants and then enter the food chain. The process of vertical migration of radioactive fallout, downwards into soil is limited. Year velocity of migration ranges from a few to several cm and depends on the type of soil, its permeability, amount of rainfall, and type, as well as properties of the element.

Moss and lichen have been used commonly as bioindicators of fallout radionuclides

[3-6]. These bioindicators have crucial advantages in determining atmospheric fallout radionuclides, such as a large surface/volume ratio, and lack of well-developed root systems. Moss has a specific surface area 10 times larger than that of herbaceous plants [7]. Another advantage is that sampling can be carried out throughout the year because of limited seasonal variations in moss and lichen morphology. Lichens grow very slowly, thus making them suitable for determining long-term deposition of fallout radionuclides.

Horwitz and his co-workers have shown that strontium may be efficiently extracted from Sr-Spec resin [8]. Nowadays, ⁹⁰Sr separation from a large variety of samples like food, milk, soil, etc. by using Sr-Spec resin is reported in the literature [9-17].

The aim of this work was to get information about ⁹⁰Sr contamination of the environment by using soil and moss from selected areas in Jeju Island, Korea. Additionally, we tried to improve the Sr-Spec method by searching for a procedure to get pure ⁹⁰Sr spectra. This belongs in the methods available as Sr-Spec resin in prepacked columns. The elution containing ⁹⁰Sr and the inactive Sr carrier was mixed with a scintillation cocktail and measured by liquid scintillation counting.

Materials and Methods

1. Materials and apparatus

All chemicals used are of analytical grade. The carrier solution Sr(NO₃)₂ is prepared by weighing from the corresponding salt. The soil and moss samples were selected from a collection of samples gathered from the Jeju Island. Reagents used for the digestion of the moss samples were 37% analytical grade HCl (Merck, Darmstadt, Germany) and 65% analytical grade HNO₃ (Merck, Darmstadt, Germany). Sr-Spec

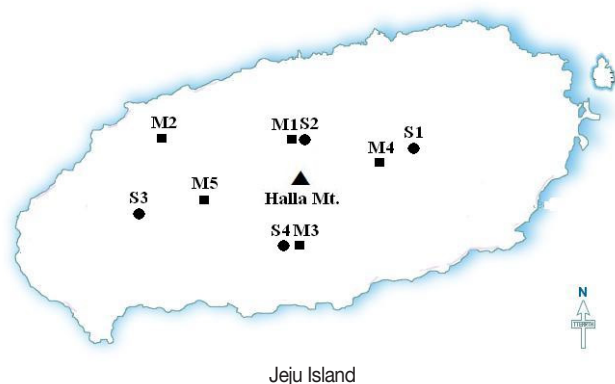


Fig. 1. Distribution of the sampling locations in Jeju Island.

resin (particle size: 100 μm to 150 μm) was obtained from Eichrom Industries, Inc. (Lisle, IL). A standard solution of ⁹⁰Sr/⁹⁰Y, provided by Korea Research Institute of Standards and Science (KRISS), was used for calibrating the counting efficiency of ⁹⁰Sr and ⁹⁰Y as a function of quenching. Separation of ⁹⁰Sr and ⁹⁰Y was carried out with extraction chromatography utilizing a Sr-Spec column. Ultima Gold LLT (Perkin Elmer, Waltham, MA) scintillation cocktail was mixed with the sample solution in a 20 mL low-potassium glass vial. Quantulus 1220 (Wallac, Turku, Finland; now Perkin Elmer, Waltham, MA) was used for the measurement of radiostrontium by liquid scintillation counting (LSC).

2. Sampling

This study took place at Jeju Island, ca. 1,400 km southeast of the Fukushima Daiichi Nuclear Power Plant. Annual rainfall of Jeju Island ranges between 1,000 mm and 1,800 mm, with an average of 1,460 mm. The altitude of Mt. Halla (center place of Jeju Island) is 1,950 m. Figure 1 shows the geographic distribution of the sampling locations in Jeju Island.

The soil samples were collected from 4 locations in 2013 (Figure 1, Table 1). Sampling area and locations are presented in Table 1. The sampling locations were selected based on the following preferences: the land should be exposed to open air without any growing tree except for small grasses, and it should be flat and stable with very little disturbance induced by natural and human actions. Our soil samples stem from the eastern part (Sangumburi, 424 m), northern part (Gwaneumsa, 548 m), the western part (Dolohreum, 692 m) and the southern part (Miaksan, 472 m) of the Mt. Halla in Jeju island. At each location, the soil sample was collected from a square area of 50 × 50 cm after removal of organic matters such as leaves or twig pieces lying on the ground. A rectangular soil core (15 × 10 cm, 20 cm depth) was taken with the help of a steel coring tool. The cored sample was carefully divided into 10 depth subsamples in every

Table 1. Geographical Information of the Soil Sampling Locations

Location	Geo code	Altitude (m)	Sampled date
S1 (Sangumburi)	33°26'8.1"N 126°41'19.2"E	424	2013-06-05
S2 (Gwaneumsa)	33°25'27.8"N 126°33'24.2"E	548	2013-07-11
S3 (Dolohreum)	33°24'18.4"N 126°27'59.6"E	692	2013-07-18
S4 (Miaksan)	33°18'14.0"N 126°33'33.0"E	472	2013-08-08

Table 2. Geographical Information of the Moss Sampling Locations

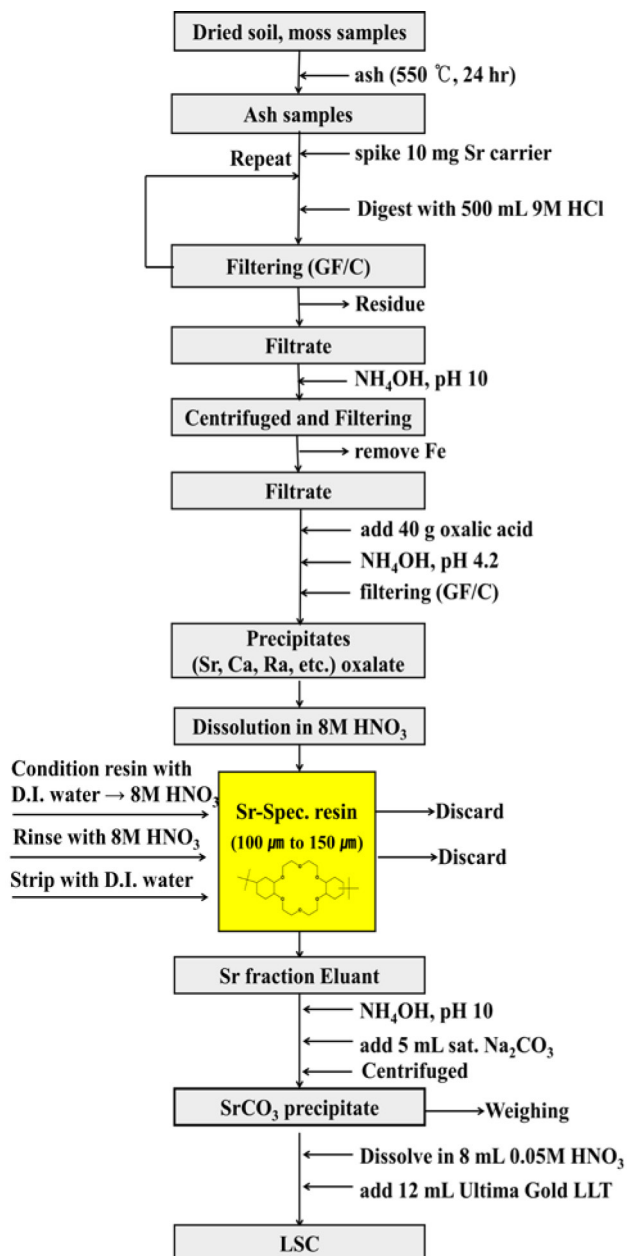
Location	Geo code	Altitude (m)	Sampling date
M1 (Gwaneumsa)	33°25'28"N 126°33'24"E	569	2011-11-16
M2 (Gwangnyeong)	33°27'38"N 126°24'38"E	122	2011-12-29
M3 (Donnaeko)	33°17'98"N 126°35'14"E	245	2012-4-12
M4 (Jeju racehorse Fostering Farm)	33°23'54"N 126°41'21"E	423	2012-5-8
M5 (1100 Meter-high Rest Area)	33°22'22"N 126°28'31"E	1076	2012-6-8

2 cm. In order to remove moisture, the soil sample was dried for 24 h in an oven kept at 105°C. The dried soil was then passed through a sieve with 2 mm holes, and pulverized for a homogeneous granularity.

The moss samples were collected from 5 locations in 2011 and 2012 (Figure 1). Sampling area and locations are presented in Table 2. The moss sample was collected by scraping it off stones or rocks existing nearby. The raw soil sample collected was weighed after removal of rubble sand organic matters to ensure that similar amounts of samples had been collected at all locations. Each moss sample was carefully cleaned from soil and other debris in the laboratory. The samples were rinsed in water twice to remove any remaining debris or soil particles. All samples were dried at 105°C for 24 hours. The samples were homogenized and powdered in the laboratory.

3. Strontium separation of soil

To determine the typical radiometric and gravimetric yields, carriers were spiked into the samples. The samples were then digested with HCl. The analysis flowchart is shown in Figure 2. The 100 g dried soil samples were ashed at 550°C in an electric furnace. Soil ashes were dissolved in 9 M HCl (500 mL), and 10.0 mg·mL⁻¹ Sr carrier (1.0 mL) was added. After stirring for 5 h, the sample solution was filtered; the beaker and filter were washed with 9 M HCl (500 mL) and a few milliliters of hot water. Iron precipitate was removed from the alkaline solution (> pH 10) by conc. NH₄OH. A few milliliters of HCl (< pH 1) and oxalic acid (40 g) were added to the solution. Strontium oxalate was precipitated by conc. NH₄OH (ca. pH 4.2). The precipitate was filtered through a GF/C filter paper. The precipitate was dissolved in 8 M HNO₃ (ca. 30 mL). Subsequently ⁹⁰Sr was purified on Sr-Spec resin. After washing with distilled water (50 mL), the Sr-spec column was preconditioned with 8.0 M HNO₃ (20

**Fig. 2.** The flowchart of procedure for isotopic analysis of ⁹⁰Sr in soil and moss samples.

mL). Then the sample solution was loaded onto the column. After rinsing with 20 mL of 8.0 M HNO₃ solution, strontium was stripped with 20.0 mL of distilled water (or 0.05 M HNO₃) and collected in a glass vial. The flow rate for all procedures was kept at 1 mL·min⁻¹.

4. Strontium separation of moss

The whole of each sample was ashed at 550°C in an electric furnace. The mass of ashes from the samples was in the

range 5 to 20 g depending on sample size. Moss ashes were dissolved in 250 mL of 9 M HCl and 0.1 mL of 10.0 mg·mL⁻¹ Sr carrier was added. After stirring for 5 hours, the sample solution was filtered; the beaker and filter were washed with a few milliliters of 9 M HCl. 20 g of oxalic acid was added to the solution. Strontium oxalate was precipitated by conc. NH₄OH (ca. pH 4.2). The precipitate was filtered through a GF/C filter paper.

The precipitate was dissolved in 8 M HNO₃ (ca. 30 mL). Subsequently ⁹⁰Sr was purified on Sr-Spec resin. After washing with distilled water (50 mL), the Sr-spec column was preconditioned with 8.0 M HNO₃ (20 mL). Then the sample solution was loaded onto the column. After rinsing with 20 mL of 8.0 M HNO₃ solution, strontium was stripped with 20.0 mL of distilled water (or 0.05 M HNO₃) and collected in a glass vial. The flow rate for all procedures was kept at 1 mL·min⁻¹.

5. Radioactivity determination

A saturation Na₂CO₃ (5 mL) was added to the stripped solution of Sr-Spec resin in the alkaline solution (> pH 10), and then allowed to stand at 50°C until a white precipitate (SrCO₃) separated. This was collected by centrifugation, and thoroughly dried *in vacuo*. The Sr yield of the respective soil sample was measured by gravimetric method. The dried sample was dissolved in 8.0 mL of 0.05 M HNO₃. Then 8.0 mL of this solution transferred to 20 mL low-potassium glass vial and 12 mL of Ultima Gold LLT scintillation cocktail; to make a total volume of 20 mL. The vial was shaken vigorously and immediately counted by LSC on a Quantulus1220. The lower limit of detection (LLD) of ⁹⁰Sr was calculated according to [18]. It was 0.1 Bq·kg⁻¹ for soil samples (counting times and chemical recovery as with ash samples, sample mass: 100 g of dried soil).

To determine the activity of radiostrontium of a sample, the counting efficiency of the radiostrontium isotope with a quench level must be measured. This was done by counting a quench series of reference standards for each isotope. Five milliliter of ⁹⁰Sr (27.96 Bq·mL⁻¹) standard solution was added in a teflon vessel and evaporated to near dryness by heating on a hot plate at 150°C. The dried standard was dissolved in 10.0 mL of 8.0 M HNO₃, then loaded onto a Sr-spec column prepared as for the samples. After washing with 30 mL of 8.0 M HNO₃ solution, strontium was stripped with 5 × 4 mL of distilled water (or 0.05 M HNO₃) and collected in a glass vial. The flow rate for all procedures was kept at 1 mL·min⁻¹. The solution evaporated by heating on a hot plate at 100°C. The

dried sample was dissolved in 10.0 mL of 0.05 M HNO₃. Then 2.0 mL of this solution transferred to five 20 mL low-potassium glass vial and 18 mL of Ultima Gold scintillation cocktail and nitromethane (0 μL, 150 μL, 300 μL, 450 μL, 600 μL) of quenching agent were added, to make a total volume of 20 mL. The vial was shaken vigorously and immediately counted by LSC.

Results and Discussion

1. ⁹⁰Sr activity concentrations of soil

The ⁹⁰Sr activity concentrations of soil samples are shown in Table 3. The ⁹⁰Sr vertical concentrations in the investigated soil samples were 18.24 ± 0.42–2.77 ± 0.22 Bq·kg⁻¹ in eastern part (S1; Sangumburi, 424 m), 18.27 ± 0.28–1.69 ± 0.11 Bq·kg⁻¹

Table 3. Activity Concentrations of ⁹⁰Sr in Jeju Island Soil Samples with Depth

Site	Depth (cm)	⁹⁰ Sr (Bq kg ⁻¹ -dry)		Ref.
		Activity	MDA	
S1	0-2	18.24±0.42	0.33	Here
	2-4	14.51±1.24	2.18	
	4-6	7.72±0.51	0.8	
	6-8	7.39±0.56	0.93	
	8-10	2.77±0.22	0.39	
S2	0-2	18.27±0.28	0.21	Here
	2-4	13.48±0.24	0.2	
	4-6	6.22±0.19	0.24	
	6-8	7.51±0.17	0.18	
	8-10	1.69±0.11	0.21	
S3	0-2	13.46±0.67	0.91	Here
	2-4	45.27±2.60	3.83	
	4-6	7.86±1.11	2.3	
	6-8	4.56±0.29	0.45	
	8-10	3.76±0.19	0.26	
S4	0-2	8.70±0.59	0.94	Here
	2-4	1.09±0.14	0.29	
	4-6	3.64±0.20	0.29	
	6-8	<MDA	2.14	
	8-10	1.46±0.41	0.96	
JeJu	0-5	8.06		[19]
	5-10	6.85		
	10-15	4.57		
	15-20	4.65		
	20-25	3.4		
Goseong	25-30	2.79		[19]
	0-5	4.57		
	5-10	4.28		
	10-15	3.82		
	15-20	3.48		
	20-25	3.58		
	25-30	3.05		

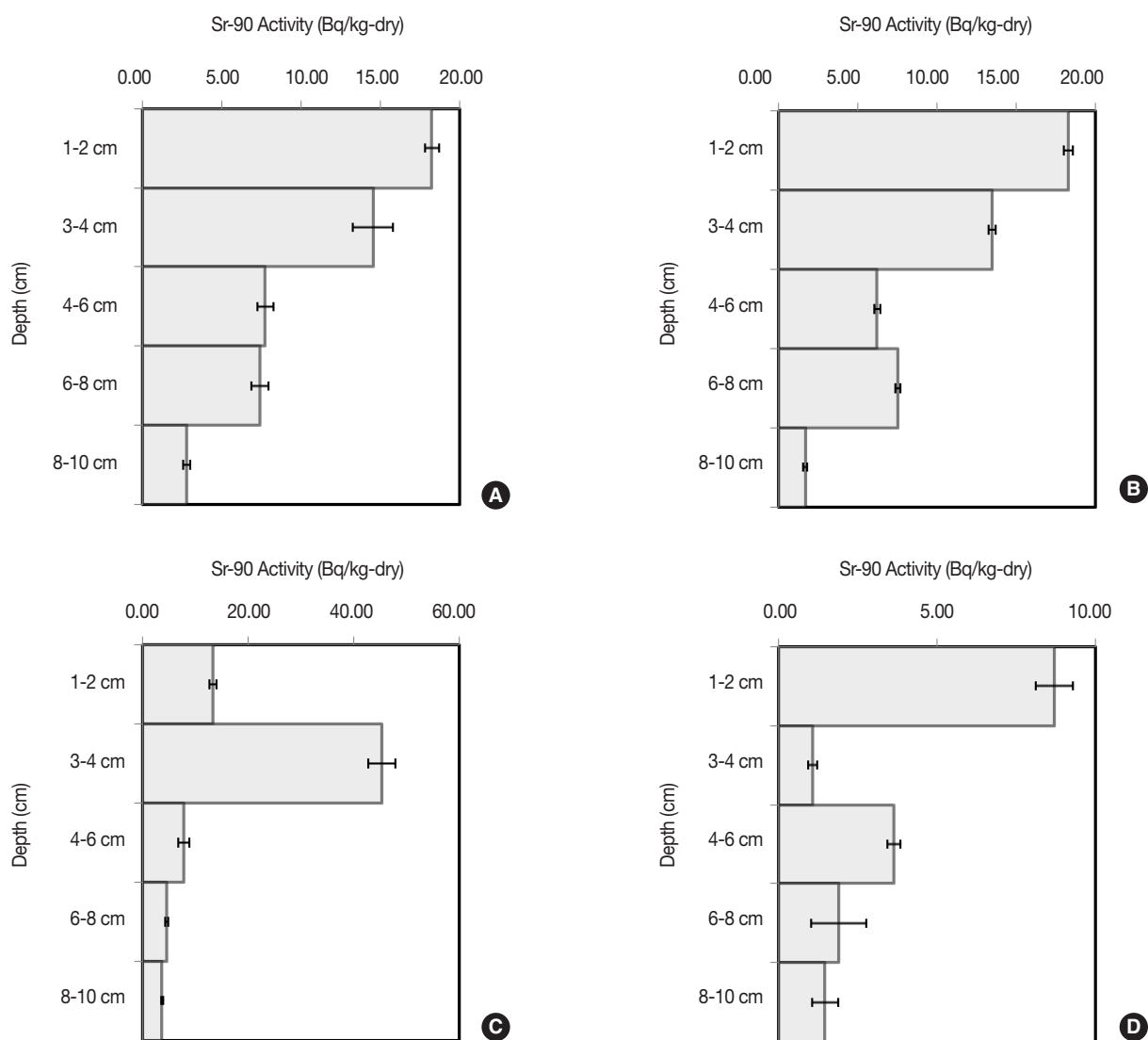


Fig. 3. Vertical distribution of ^{90}Sr activities in the soils from Jeju island: (A) S1, (B) S2, (C) S3, and (D) S4 sites.

in northern part (S2; Gwaneumsa, 548 m), 45.27 ± 2.60 - $3.76 \pm 0.19 \text{ Bq}\cdot\text{kg}^{-1}$ in the western part (S3; Dolohreum, 692 m) and 8.70 ± 0.59 - $1.09 \pm 0.14 \text{ Bq}\cdot\text{kg}^{-1}$ in the southern part (S4; Miaksan, 472 m) of the Mt. Halla in Jeju island, respectively.

Activities of ^{90}Sr show the highest value at the surface soil and decrease with depth (Figure 3).

2. ^{90}Sr activity concentrations of moss

Moss samples were measured immediately after extraction, one week later and finally after two weeks. Samples were spiked prior to digestion to correct for losses of analytes during sample preparation procedure. The Sr concentrations in the investigated mosses samples were $363 \pm 3.0 \text{ Bq}\cdot\text{kg}^{-1}$, $97.7 \pm 1.0 \text{ Bq}\cdot\text{kg}^{-1}$, $91.7 \pm 0.7 \text{ Bq}\cdot\text{kg}^{-1}$, $79.6 \pm 0.8 \text{ Bq}\cdot\text{kg}^{-1}$,

Table 4. ^{90}Sr Activity Measured with Liquid Scintillation Counting in Moss Samples from Jeju Island

Site	^{90}Sr ($\text{Bq}\cdot\text{kg}^{-1}$ -dry)	
	Activity	MDA
M1	363.00 ± 3.0	0.98
M2	97.70 ± 1.0	0.50
M3	91.70 ± 0.7	0.26
M4	79.60 ± 0.8	0.37
M5	225.00 ± 2.0	0.64

and $225 \pm 2 \text{ Bq}\cdot\text{kg}^{-1}$ (-dry weight), respectively. The chemical yield and the ^{90}Sr activity concentration of each sample are shown in Table 4. The measured ^{90}Sr concentration is higher at high-altitude locations.

Conclusions

In this study, we determined ⁹⁰Sr concentrations in soil and moss of Jeju Island (Korea) using strontium specific ion exchange columns in combination with liquid scintillation spectrometry. A large amount of airborne radioactive species had been released following the Fukushima Daiichi nuclear reactor accident. We present an efficient method of measuring the ⁹⁰Sr activity in environmental samples, which is based on using ion exchange columns loaded with a Sr-Spec resin and liquid scintillation spectrometry. The activities of ⁹⁰Sr in soil and moss samples were investigated at nine locations of Jeju Island, Korea. The ⁹⁰Sr vertical concentrations in the investigated soil samples were 18.24 to 2.77 Bq·kg⁻¹ in eastern part, 18.27 to 1.69 Bq·kg⁻¹ in northern part, 13.46 to 3.76 Bq·kg⁻¹ in the western part and 8.70 to 1.09 Bq·kg⁻¹ in the southern part of the Mt. Halla in Jeju island, respectively. Activities of ⁹⁰Sr show the highest value at the surface soil and decrease with depth. The measured activity concentrations were between 79.6 Bq·kg⁻¹ -dry (moss in Jeju racehorse Fostering Farm site) to 363 Bq·kg⁻¹ -dry (moss in Gwaneumsa site). The result will be first study of the activity of ⁹⁰Sr in moss samples from Jeju Island, Korea.

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