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■論 文■

Cumulative Deposition of ^{137}Cs in the Soil of Korea

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한국토양에 존재하는 ^{137}Cs 방사능 분포

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Abstract - The cumulative deposition of ^{137}Cs in the soil of Korea has been studied. Using γ-ray spectrometry, the concentrations of ^{137}Cs were determined for the soil samples collected to a depth of 20 cm. The average accumulated depositions of ^{137}Cs were estimated roughly to be 2,501 ± 499 Bq m^{-2} in the forest and 1,058 ± 322 Bq m^{-2} in the hill. The inventory value of ^{137}Cs in the forest is about two times higher than that in the hill. Except for some cases, the concentrations of ^{137}Cs in the undisturbed soils decreased exponentially with increasing the soil depth. The influences of rainfall, organic matter content, clay content and pH on the deposition of ^{137}Cs were studied using the field method. Among these factors, the organic matter content played the most important role in the retention and relative mobility of ^{137}Cs in the soil. The other factors such as rainfall, clay content and pH showed weak correlation with the deposition of ^{137}Cs in the soil.
Key words : ^{137}Cs, cumulative deposition, organic matter content, clay content, pH

요약 - 한국 토양에 존재하는 ^{137}Cs 방사능 분포에 관한 연구를 수행하였다. ^{137}Cs 방사능 농도를 결정하기 위하여 남한지역 17개 지점에 대하여 20㎝ 깊이로 토양을 채취한 후, 감마스펙트로메타를 이용하여 ^{137}Cs 방사능 농도를 결정하였다. 야산지역과 숲속에 대한 ^{137}Cs 의 평균 농도는 각각 1,058± 322 Bq m^{-2} 및 2,501±499 Bq m^{-2} 로, 숲속의 ^{137}Cs의 농도가 야산지역보다 약 두배정도 높은 수치를 보였다. 인위적 교란이 없는 지역에서 토양깊이 변화에 따른 ^{137}Cs 분포는 토양깊이가 증가할수록 지수함수적으로 감소경향성을 나타내었다. ^{137}Cs 농도 분포에 영향을 미치는 여러인자를 조사한 결과 유기물 함량이 ^{137}Cs의 토양 침적에 중요한 작용을 함을 알 수 있었고 강수량, 점토의 함량 및 pH과 같은 인자들은 ^{137}Cs의 토양 침적과 상관관계가 거의 없습을 발견 할 수 있었다.
중심어 : ^{137}Cs, 농도분포, 유기물함량, 강수량, 점토함량, pH

INTRODUCTION

Worldwide radioactive contamination in the soil was caused by the major nuclear weapon tests conducted in the late 50s and early 60s. Large scale contamination has resulted from the Chernobyl accident of 1986. Most of the radioactive elements produced by atmospheric nuclear tests decay so rapidly that a few elements with long half-lives may remain in the soil at the present time. Among the artificial radionuclides released into the environment, ^{137}Cs is a good tracer for studying the behavior of these radionuclides in the environment, because of its high radiotoxicity, long half-life and long residence in biological systems. The contamination level of ^{137}Cs in the terrestrial environment due to atmospheric fallout is highly variable with time and geographic location. Cesium-137 is strongly adsorbed on clay minerals such as mica and kaolin and its downward migration is limited. Since cesium-137 is a gamma-emitter, it is possible to measure its concentration in environmental samples with high accuracy without chemical separation. With its long half-life (30.2 yr.) and the total fallout deposited especially in the northern hemisphere,

Table 1. Inventory of ^{137}Cs in soils and other environmental data.

Location	Type of Vegetation	Organic matter Content(%)	Rainfall(mm y^{-1})	Texture(%) Clay	Silt	Sand	pH	Inventory of ^{137}Cs(Bq m^{-2})
YANGGU	Hill	6.0	1,296	10.2	50.4	39.4	4.7	1450
ULJIN	Forest	12.2	1,190	12.4	15.6	72.0	4.9	2447
KANGLUNG	Forest	10.8	1,580	15.9	27.2	56.9	5.0	2180
JUNGSUN	Forest	11.0	1,357	17.3	43.7	39.0	4.8	2571
EUIWANG	Forest	11.5	1,307	19.3	27.9	52.8	4.3	3205
YEIJU	Hill	5.7	1,304	9.4	25.0	65.6	4.7	1374
BOEUN	Forest	6.8	1,250	9.0	19.2	71.8	5.5	2463
GONGJU	Forest	13.7	1,311	17.3	48.6	34.1	4.7	3086
TAEJON	Arable	4.9	1,260	13.6	8.6	78.9	4.0	738
SESAN	Hill	4.8	1,216	16.8	40.2	43.0	4.4	869
ANDONG	Hill	7.5	1,274	16.3	21.4	62.3	5.0	772
KORUNG	Hill	8.0	1,011	14.2	32.8	53.0	4.5	1345
KEINGJU	Hill	12.4	1,272	15.4	25.1	59.5	4.7	1023
KIMJE	Forest	10.6	1,280	15.8	30.8	53.4	4.3	2447
KOCHANG	Forest	9.4	1,249	12.6	10.4	77.0	5.1	1612
HAEHAM	Hill	4.2	1,360	15.6	29.4	55.0	4.6	538
HADONG	Hill	6.4	1,301	13.4	22.3	64.3	4.9	1095

^{137}Cs will be detectable in environmental samples for many years. Several studies on the distribution of ^{137}Cs were carried out in the soil of Korea[1-3]. However, these results on the concentration of ^{137}Cs were limited to only surface soil (0~5 cm). Therefore, it is impossible exactly to clarify the cumulative deposition of ^{137}Cs in the soil.

The purpose of this study is not only to establish base-line data on the cumulative deposition of ^{137}Cs in the Korean soil due to the global nuclear weapon test, but also to investigate the effect of organic matter content, rainfall, clay content and pH on the deposition of ^{137}Cs in the soil.

MATERIALS AND METHODS

Soil sampling and preparation

A sampling area is very important in the determination of ^{137}Cs contamination level in the terrestrial environment due to atmospheric fall-out. In the field method, soil samples were collected from 17 sites in South Korea. All the sampling points were selected on a flat area, in order to minimize the possibility of precipitation run-off. Soil sampling was carried out from 1994 to 1995. At each site, two soil samples were taken with a soil sampler within an area of about 30 ㎝ x 30 ㎝ to the depth of about 20 ㎝. Several preliminary experiments on forest soils showed that a sampling depth of 20 ㎝ is sufficient to account for more than 95 % of deposited activities of radionuclides from global fallout[2-3]. Samples were dried at 110℃ for 48h after removing pebbles and fragments of plant root from the samples, and then sieved through a 1.0 mm screen.

Analytical procedures

Organic matter content in the samples was determined using the loss-on-ignition method. The samples were ashed at 550℃ for 24h[4]. The soil texture was analyzed using a Hydrometer method[5]. The pH was measured with a glass electrode in a 1: 5 suspension of soil and water[6].

For the γ-ray spectrometry, soil samples were oven-dried at 110℃ to reach a constant weight. A HPGe detector with the counting efficiency of 25 % and FWHM of 1.7 keV was used. The detection limit was 0.2 Bq kg^{-1} soil for 72,000s counting time. The counting efficiency was calibrated by using the mixed sources (Amersham, UK) in suitable substrates and geometries. The corresponding γ-ray spectra were analyzed using the Omnigam Software.

RESULTS AND DISCUSSION

Inventory of ^{137}Cs

In Table 1, the cumulative deposition of ^{137}Cs

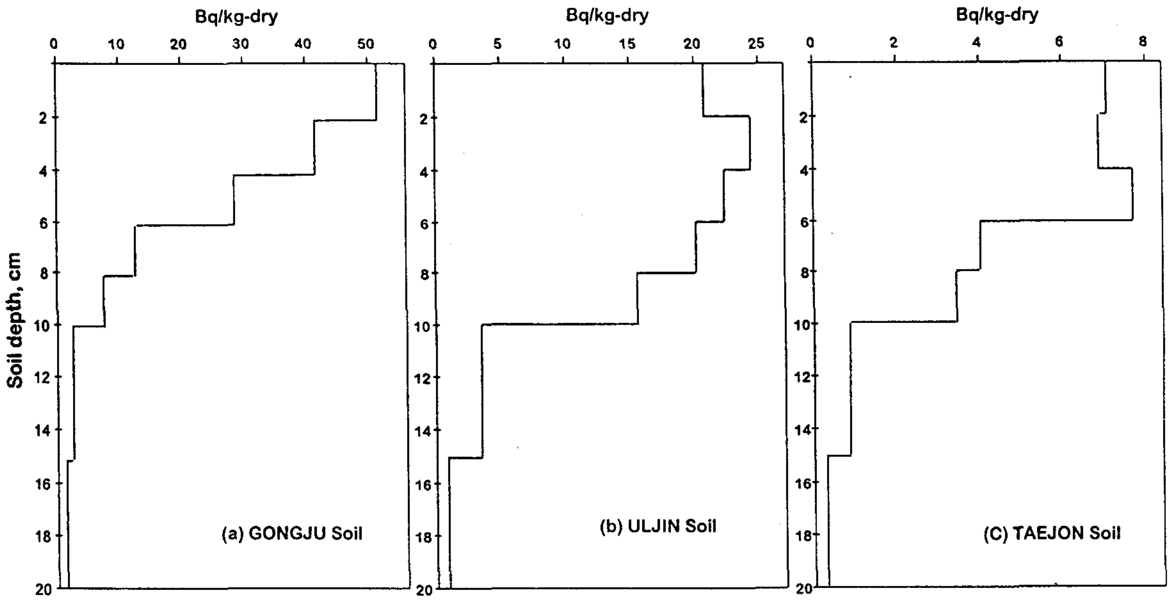

Fig. 1. Depth profiles of ^{137}Cs in Soils.

in the soil samples were summarized in terms of activities per unit area (Bq m^{-2}). The inventory represented as Bq m^{-2} is much more meaningful than the observed activity concentrations (Bq kg^{-1}), because the observed activity varies with parameters such as soil type, topography, water content, mineralogy and rate of soil erosion. As presented in the Table 1, the deposited activities of ^{137}Cs were estimated in the range of 538 to 1,450 Bq m^{-2} with a average value of 1,058±322 in the soils of hill. In the soils of forest, the accumulated depositions of ^{137}Cs were estimated in the range of 1,612 to 3,205 Bq m^{-2} with a average value of 2,501±499. These inventory values are lower than those in Europe affected by Chernobyl accident[7]. The ^{137}Cs inventory in Japanese soil in 1980 was reported to be distributed from 3,000 to 4,700 Bq m^{-2}[8], which is higher than those in this study. This difference may suggest that the deposition rate can be influenced by meteorological condition and topographical features of the earth surface, such as soil type, organic substances or clay content. Also, the average inventory of ^{137}Cs in the forest is two times higher than that in the hill. It means that the fallen leaves in the deep forest accumulate fallout radionuclides in the nearby soil. The deciduous leaves of a tall tree on the ground increase not only the effective surface area for deposition of fallout but also the amount of humic substances which may easily associate with the fallout radionuclides.

Depth profile of ^{137}Cs

After their deposition of fallout radionuclides on the soil surface, they migrate downward. The depth profile of the fallout radionuclides can give information on the retention and migration of the radionuclides.

The depth distribution of ^{137}Cs in top 20 cm soil layer for typical three sites was shown in Fig. 1. As shown in Fig. 1-a, the concentration of ^{137}Cs in the grassland and forest soil decreases exponentially as the depth of soil increases. Most of ^{137}Cs is accumulated in the layer of upper 10 ㎝ of the soil. The contribution of the upper layer to total inventory of ^{137}Cs were more than 90 %. It implies that some inorganic substances in the surface soil, particularly clay minerals such as mica and kaolin, have a large capacity of adsorbing and fixing Cs and thereby limit downward migration [9, 10]. However, somewhat different pattern in the depth profile was observed in the ULJIN soil as shown in Fig. 1-b. The maximum concentration of ^{137}Cs was found not in the top layer but in the second layer of 2-4 ㎝ depth.

This may be explained either by the downward percolation through the soil due to the rainwater or by the outflowing loss of the fine particle fractions adsorbing higher amounts of these nuclides. The similar depth profile was also observed on the KOHNO area in Japan [11]. In Fig. 1-c, concentrations of ^{137}Cs in the cultivated soil around TAEJON area were significantly lower than those found in the forest soils studied in this work and constant up to the depth of 8-10 cm below. The result is probably due to the effect of tillage carried out in these plots. The tillage, mixing the soil, must have disturbed the distribution of ^{137}Cs in the whole profile.

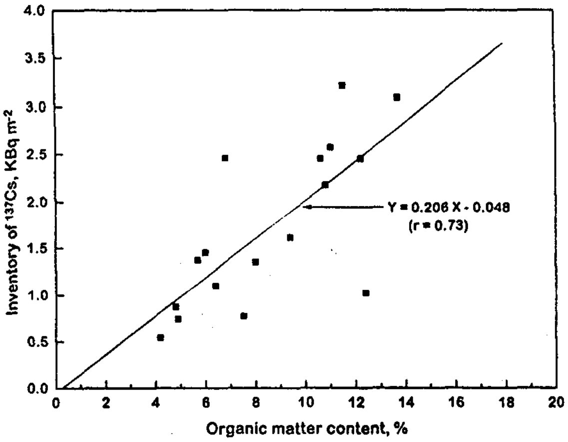

Fig. 2. The correlation between concentrations of ^{137}Cs and organic matter content.

Factors affecting cumulative deposition of ^{137}Cs

The large number of factors such as organic substances, rainfall, clay content and pH affects deposition of ^{137}Cs. The contents of organic matter in the surface soil samples and other characteristics of the soil were shown in Table 1.

Regression analysis was used to investigate a possible correlation between the concentration of the radionuclides and the content of organic matter in the soil. As shown in Fig. 2, the correlation between the organic matter content and the concentration of ^{137}Cs is highly significant, and its correlation coefficients was 0.71. Owing to its large cation exchange capacity, the organic matter content is a characteristic factor that has a significant influence on the dynamics of fallout-originated radionuclides in the soil [12-14]. Thereby, it must be certain that ^{137}Cs easily associates with the organic substances as forms of surface coating and complexing. Higher concentration of ^{137}Cs found in the soil with high organic content must be attributed to these phenomena.

Rainfall (including snowfall) has been known to influence deposition of ^{137}Cs by worldwide

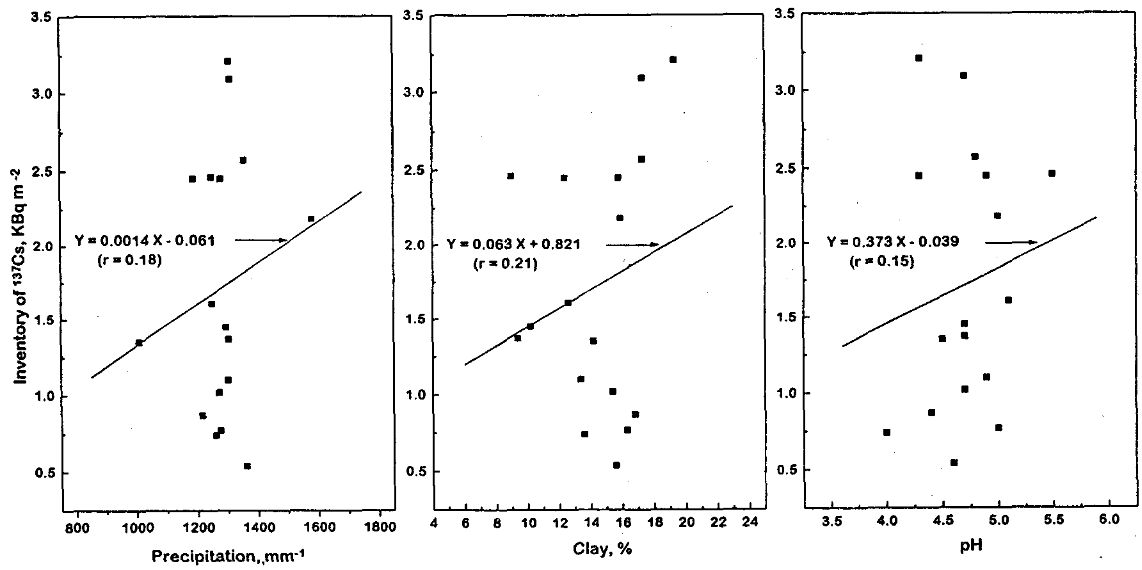

Fig. 3. The correlation between concentrations of ^{137}Cs and precipitation(a), clay percentage(b) and pH(c).

fallout, and could be used as an indicator of the distribution of these nuclides in soil[15]. Precipitation scavenging is an important mechanism which brings the debris of nuclear explosions in the atmosphere to the earth surface. The accumulated deposit depends mainly on rainfall and latitude[16]. Krey and Beck[17] reported that areas in Utah receiving higher rainfall showed higher levels of fallout radionuclides than the areas with lower rainfall. To deduce the relationships between the concentration of ^{137}Cs and the rainfall, linear correlation analysis was carried out. The results were shown in Fig. 3-a. The correlation coefficient was 0.18, which is much less than that of organic matter content. The weak correlation may be explained by either the outflowing loss of the fine particle fraction adsorbing the radionuclides by heavy rain in the rainy season, or partial soil erosion by severe temperature difference between summer and winter. Since the amount of rainfall is more or less constant in Korea, it is difficult to deduce the correlation between the concentration of ^{137}Cs and rainfall.

The clay content in the soil may affect the concentration of fallout radionuclides. It is expected that the activity concentration of fallout radionuclides in the soil increases with the increase of clay content, as a consequence of the greater adsorption capacity and the increased surface area. However, no significant correlation between the clay content and the concentration of ^{137}Cs in the soil has been observed (r = 0.21), as presented Fig. 3-b. This observation implies that the physical properties of the soil, such as soil texture, is less affecting the deposition of ^{137}Cs than organic matter content.

The pH value affects adsorption and solubilization processes of radionuclides in soil. By means of a linear correlation analysis, the influence of the pH on the ^{137}Cs concentration in the soil was studied. As shown in Fig. 3-c, the correlation between the ^{137}Cs concentration and the pH values was not significant (r = 0.15). The weak effect of pH on the distribution of radionuclides has been reported in the previous literature: Schimmack *et al.*[18] has observed no pH effect on the vertical distribution of Cs in soil by an experiment carried out by adding water at different pH. The weak correlation may imply that pH has no influence on the retention of ^{137}Cs in the soil in the given range of pH, or the pH effect is masked by the action of the other parameters affecting ^{137}Cs mobility in the soil.

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SUMMARY

In this study, the concentrations of ^{137}Cs were determined for the soil samples collected to a depth of 20 cm using γ-ray spectrometry. The average values of accumulated depositions of ^{137}Cs in the soil were estimated roughly to be 2,501 $\pm$ 499 Bq m^{-2} in the forest and 1,058 $\pm$ 322 Bq m^{-2} in the hill. The inventory value of ^{137}Cs in the forest is about two times higher than that in the hill. Except for some cases, the concentrations of ^{137}Cs in the undisturbed soils decreased exponentially with increasing the soil depth. The influences of rainfall, organic matter content, clay content and pH on the deposition of ^{137}Cs were studied using the field method. Among these parameters, the organic matter content played the most important role in the retention and relative mobility of ^{137}Cs in the soil. The other factors such as rainfall, clay content and pH showed weak correlation on the deposition of ^{137}Cs in the soil.

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