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ORIGINAL ARTICLE

The contents and forms of solid-phase species of radioactive strontium and cesium in Taiwan soils

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Abstract This study was to investigate the activities and contents of ^{137}Cs in the profiles of selected arable and forest soils in Taiwan and various solid-phase species of ^{85}Sr and ^{137}Cs in selected arable soils in Taiwan. The gamma (γ) ray spectra of the collected soil samples and some of the soils amended with ^{85}Sr and ^{137}Cs were measured. The data indicate that the arable soils from Sanhsing series, Sanhsing Township and Chuangwei series, Chuangwei Township, Ilan County, and from Tunglochuan series, Pinglin Township, Taipei County shows significantly higher radioactivity of ^{137}Cs (ND $-$ 11.0 $\pm$ 0.2 Bq kg^{-1}). Furthermore, the radioactivity of ^{137}Cs in the mountain soils (1.24 $\pm$ 0.07 $-$ 42 $\pm$ 1 Bq kg^{-1}) from Yuanyang Lake Nature Preserve among Ilan, Taoyuan, and Hsinchu Counties is the highest among the investigated mountain forest soils. This may be mainly attributed to the fact that Ilan County is located in the northeastern part of Taiwan and faces the northeastern and northern seasonal winds with lots of precipitation annually from mid-autumn through mid-spring next year and is receiving greater amount of fallouts yearly. Due to longer reaction period (>3 y) of ^{137}Cs with soil components, ^{137}Cs was mainly in the forms bound to oxides and to organic matter in the soil amended with ^{137}Cs and in the soil contaminated with ^{137}Cs. On the contrary, due to shorter reaction period (<60 d) of ^{85}Sr with soil components, ^{85}Sr was mainly in exchangeable form and partially in the forms bound to carbonates and oxides in the soils amended with ^{85}Sr.

Keywords ^{137}Cs · ^{85}Sr · Solid-phase species · Gamma (γ) ray spectra · Radioactivity

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1 Introduction

Radionuclides are unstable isotopes, which undergo radioactive decay. Some occur naturally in air, rocks, soils and plants at concentrations that give measurable amounts of radiation and some are produced artificially, as in nuclear weapon testing. Radiation of alpha (α), beta (β) and gamma (γ) rays are from radioactive decay of radio nuclides. All these kinds of radiation are a health hazard. The hazard is greatest if the radionuclide enters the body through the intake of contaminated food, water, or air. Radionuclides of cesium and strontium have long half-lives and although they are adsorbed by soils (cesium is only weakly adsorbed by peat) they can pass into the food chain, where they represent a hazard to grazing animals and humans (Wild, 1993). A literature survey of bomb fallout titles from 1970 to the present revealed that only the following radioisotopes were cited a significant number of times: $^{239-240}$Pu, ^{36}Cl, ^{129}I, ^{90}Sr, and ^{137}Cs. Moreover, radioactive fallouts from above ground nuclear weapon testing and nuclear facility accident releases are the primary source of ^{90}Sr and ^{137}Cs in soil (Zhang *et al*., 2002). In fallouts, the long half-lived ^{90}Sr (29 yr) and ^{137}Cs (30.2 yr) are two of the most radioactive radionuclides (Hsu, 1996). Thereafter, depending on residence time in soil environment and chemical characteristics, these radionuclides may infiltrate to deeper soil layer with rainfall and irrigation water. Consequently, they may be absorbed by plant roots or be leached by water down to much deeper layer to incorporate into groundwater (Bunzl *et al*., 1995; Zhang *et al*., 2002). The chemical behaviors of ^{90}Sr and ^{137}Cs are close to those of the plant essential elements of Ca and K, respectively. ^{90}Sr and ^{137}Cs thus directly and indirectly threaten ecological receptors as well as human beings, the ending-member of food chain (Lai *et al*., 1997). Additionally, radionuclides may be absorbed by plants, microorganisms, fungi or animals, and then enter human bodies by bio-chain (Holm, 1994). The target organs of ^{90}Sr and ^{137}Cs are human being's bone marrow and the whole body, respectively (Zhang *et al*., 2002). High energy of ionized radiation released from radionuclides may destruct the human cells and consequently cause human's genetic variation and cancer (Holum, 1994). The chemical behavior of ^{85}Sr is similar to that of ^{90}Sr. However, the half-life of ^{85}Sr (64.7 d) (Zhang *et al*., 2002) is much shorter than that of ^{90}Sr (29 yr) (Hsu, 1996). Further, the measurement of radioactivity of gamma (γ) ray spectrum of ^{85}Sr by using a gamma spectrometer equipped with a high purity germanium semiconductor, Ge (Canberra GC4020) is much more accurate and time-saving than the measurement of radioactivity of β emission of ^{90}Sr (Taiwan Radiation Monitoring Center, 1993). This study was aimed to investigate (1) the contents and distributions of radioactive ^{137}Cs in the profiles of selected arable soils in Taiwan, and (2) the effect of soil properties on the distributions of solid-phase species of ^{85}Sr and ^{137}Cs in the red soils and alluvial soils after being amended with ^{85}Sr or ^{137}Cs.

2 Materials and methods

2.1 Collection of soil samples and analyses of soil properties

The representative arable soils were collected according to the Taiwan Soil Map. The selection of types of soil samples was based on the classification of major soil groups at the selected sampling sites of townships and cities. Distribution of the 17 arable soil sampling sites is shown in Fig. 1 and Table 1. The nomenclatures of the soil groups described in Table 1 are based on the reports compiled by Soil Survey Staff (1996) and Tsai *et al*. (1998). In addition, two more mountain forest soils were collected from Wuling Farm, Hopin Township, Taichung County and from Yuanyang Lake Nature Preserve, located at the border of Ilan, Taoyuan, and Hsinchu Counties (Table 1 and Figure 1).

For each soil sampling site described in Fig. 1 and Table 1, ten adequately equal amounts of the sub-samples were randomly collected from ten subsampling sites by an auger (8.3 cm in diameter) from the sketched grids of each ten-hectare area and then mixed to make a composite sample. The center location expressed as latitude/longitude of the ten subsampling sites in each ten-hectare area is shown in Table 1.

The sampled depths at Shanhua, Kuantein, and Hsuejia Townships in Tainan County were 0–15 and 15–30 cm. However, based on the consideration of the sedimentation of fallouts on soils and of their subsequent downward movement in soil profiles, and based on the consideration of the risk of absorption of radioactive ^{137}Cs by root systems of planted edible crops, all soil samples from other sites were collected from the depths of 0–5, 5–10, 10–20, and 20–30 cm. All composite soil samples were collected in triplicate from each sampled depth and composite samples were air-dried, ground, and passed through a 2-mm sieve.

From all the selected sampling sites, separate surface soils (0–15 cm) of the yellow-brown red soil of Longzhong series, alluvial soil of Changhsing series, red soil of Pinchen series, and alluvial soil of Chunliao series were additionally collected and subjected to the analyses of some soil physical and chemical properties. The properties selected for analyses are considered to be pertinent for influencing the characteristics

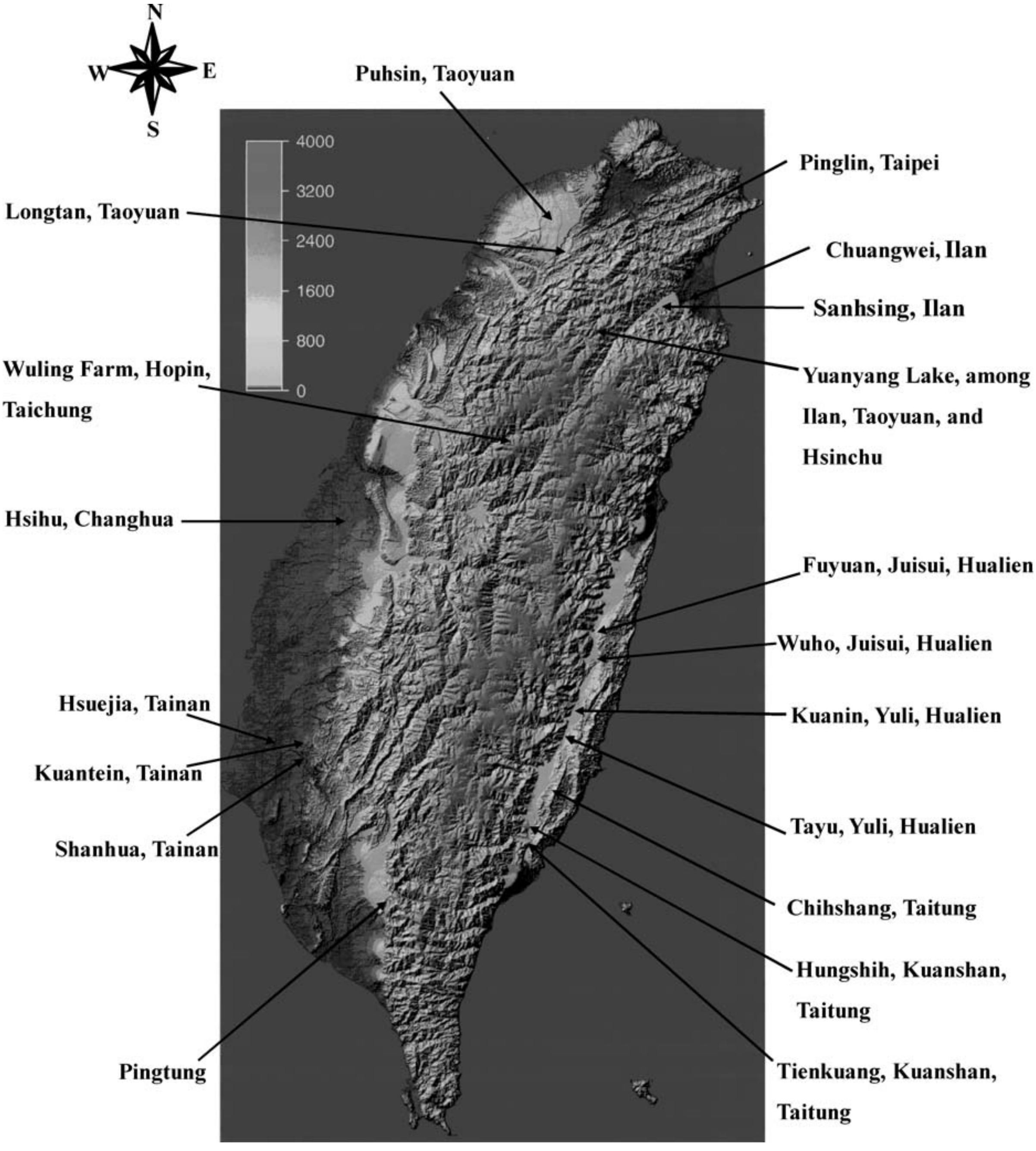

Fig. 1 Sampling locations and topography of Taiwan.

and forms of ^{85}Sr and ^{137}Cs in soil environment. The selected properties include pH (McLean, 1982), EC (electrical conductivity) (Rhoades, 1982a), organic carbon contents (Tiessen *et al*., 1981), textures (Gee and Bauder, 1986), CEC (cation exchange capacity) (Rhoades, 1982b) and effective CEC (Gillman, 1979), exchangeable Ca, Mg, K and Na (Thomas, 1982), CBD (citrate-bicarbonate-dithionite) (McKeague and Day, 1966; Jackson, 1979), oxalate (Schwertmann, 1964; McKeague and Day, 1966), and pyrophosphate (Loveland and Digby, 1984) extractable Mn, Fe and Al in oxides.

Table 1 Locations and classifications of soil samplings

Sampling site	Classification	Latitude/Longitude
Pinglin	Hapludult	2759450 mN, 317550 mE
Puhsin, Taoyuan	Kandiperox	2755850 mN, 267850 mE
Longtan, Taoyuan	Paleudult	2749250 mN, 273950 mE
Wuling Farm, Hopin, Taichung		2694550 mN, 282150 mE
Hsihu, Changhua	Psammaquent	2649550 mN, 190550 mE
Hsuejia, Tainan	Haplaquent	2572650 mN, 166550 mE
Kuantein, Tainan	Eutrochrept	2565050 mN, 180550 mE
Shanhua, Tainan	Ochraqualf	2558250 mN, 178150 mE
Pingtung	Epiaquent	2507350 mN, 198450 mE
Chuangwei, Ilan	Fluvaquent	2733350 mN, 327250 mE
Sanhsing, Ilan	Epiaquent	2729250 mN, 314350 mE
Yuanyang Lake		2719350 mN, 290150 mE
Fuyuan, Hualien	Eutrochrept	2608750 mN, 287750 mE
Wuho, Hualien	Hapludult	2596650 mN, 286650 mE
Kuanin, Hualien	Vdorthent	2587750 mN, 285350 mE
Tayu, Hualien	Vdorthent	2586550 mN, 282950 mE
Chihshang, Taitung	Vdorthent	2558350 mN, 272650 mE
Hungshih, Taitung	Vdorthent	2551350 mN, 266450 mE
Tienkuang, Taitung	Vdorthent	2547550 mN, 267350 mE

2.2 Measurements of ^{137}Cs and ^{85}Sr activity of the soils

Each of the air-dried soil samples from the 19 sampling sites was filled into a Mevell cup of 12 cm height and the gamma ray spectra was measured by using a gamma spectrometer equipped with a high purity germanium semiconductor, Ge (Canberra GC4020). The detection range of the spectrometer was from 50 to 10000 keV. The relative efficiency of the measurement was 40%. The energy value of ^{137}Cs in a gamma ray spectrum of calibrated reference source was 661.66 keV (Skoog *et al*., 1998). The radioactivity of soil samples collected from each sampling site was measured in triplicate.

2.3 Amendments of ^{85}Sr and equilibration of ^{85}Sr- and ^{137}Cs-containing soils

Due to inconvenience of the measurement of β emission from ^{90}Sr (Taiwan Radiation Monitoring Center, 1993), ^{85}Sr which has chemical behavior similar to ^{90}Sr (but convenient to measure gamma emission) was used as a tracer in the experiment. Five grams of the concentrated standard ^{85}Sr solution with the activity of 1.04 MBq g^{-1} (Damri Inc.) was diluted to 100 mL with 0.1 M HCl. The radioactivity of the diluted solution was around 3.8×10^4 Bq g^{-1}. The soil water content at field capacity of each of the four soils subjected to the analyses of soil properties (described above) was determined at 1/30 MPa tension by a pressure plate apparatus. At soil field capacity water has moved out of the macropores and air has moved in, to take its place. The micropores or capillary pores are still filled with water and can supply plants the needed water. In other words, soil water content at field capacity is the most suitable moisture regime for crop growth (Or and Wraith, 1999; Brady and Weil, 1999). The ^{85}Sr-containing solution with radioactivity of 3.8×10^4 Bq g^{-1} was diluted with double-distilled water to make ^{85}Sr solution equal to the field capacity equivalent amount of water. It was then added to 5 kg of red soil (Pinchen series) or of alluvial soil (Chunliao series) in a pot and equilibrated for 32 d. In the same manner, a field capacity equivalent amount of double-distilled water was added to 5 kg of ^{137}Cs-contaminated yellow-brown red soil (Longzhong series) and ^{137}Cs-amended alluvial soil (Changhsing series) and equilibrated for the same periods. During the equilibration period, the water contents of the four soils in the pots were maintained at their respective field capacity values by weighing.

2.4 Extractions and determinations of various forms of solid-phase species of ^{85}Sr and ^{137}Cs in the soils

At the end of the 32 d reaction period, three replicate portions from each of the ^{85}Sr-amended red soil and alluvial soil as well as ^{137}Cs-contaminated yellow-brown red soil and ^{137}Cs-amended alluvial soil as described above were collected from each pot. Each soil was subjected to the extraction of exchangeable form, forms bound to carbonates, Fe-Mn oxides, and organics, and residual form of ^{85}Sr or ^{137}Cs, according to the sequential extraction procedure recommended by Tessier *et al*. (1979). The energy value of ^{85}Sr in a gamma ray spectrum of a calibrated reference source was 514.01 keV (Skoog *et al*., 1998).

3 Results and discussion

3.1 Basic soil properties

The yellow-brown red soil (Longzhong series) and red soil (Pinchen series) were silty clay and clay, respectively, while alluvial soils (Changhsing series and Chunliao series) were loamy sand (Table 2). Soils of Longzhong series and Pinchen series were acidic whereas soils of Changhsing series and Chunliao series were slightly acidic and neutral, respectively (Table 2). These could be attributed to their different parent materials and/or degree of weathering. Soils of Longzhong series, Changhsing series, and Chunliao series had significantly higher EC values than that of Pinchen series (Table 2), showing the differences in fertilization of cultivation. According to results in Table 2, soil of Changhsing series with the texture of loamy sand has the highest CEC among the investigated soils. This can be mainly attributed to its higher organic C content. Soil of Chunliao series had the lowest organic C content among the soils. This may be attributed to its coarse texture of loamy sand with difficulty of the formation of organo-clay complex, leading to easily oxidative decomposition of organic matter. Soils of Longzhong series, Changhsing series, and Chunliao series had the highest or higher contents of exchangeable K and Na, Ca and Mg, and Ca, respectively, reflecting their characteristic pH and EC values (Table 2). The crystalline and noncrystalline forms and the form bound to organics of Fe, Al, and Mn are extractable by CBD (McKeague and Day, 1966; Jackson, 1979), oxalate (Schwertmann, 1964; McKeague and Day, 1966), and pyrophosphate (Loveland and Digby, 1984), respectively. The sequence of the contents of various forms of Fe in the soils of Longzhong series and Pinchen series was crystalline > bound to organics > noncrystalline, whereas that for the soils of Changhsing series and Chunliao series was crystalline > noncrystalline > bound to organics (Table 3). The contents of the three forms of Al in the soils of Changhsing series and Chunliao series were lower than those of their corresponding forms of Fe (Table 3). The contents of the three forms of Mn in the four soils were the least among their corresponding forms of Fe and Al (Table 3).

3.2 The radioactivity, contents, and distribution of ^{137}Cs in the soils

As seen from Figs. 1 and 2, in the south of this island, the surface soil samples (0–15 cm in depth, selected based on the consideration of soil-root-crop relationship) collected from three sites in Tainan County, the range of radioactivity of ^{137}Cs was from 0.62 ± 0.7 to 2.0 ± 0.2 Bq kg^{-1} (not shown), which is higher than the average (0.5 Bq kg^{-1}) of general typical soil samples (Lai *et al*., 1995). In order to investigate the possible sedimentation of fallouts and their subsequent downward movement in soil profile, the remaining soil samples from other sampling sites were collected from the depths of 0–5, 5–10, 10–20, and 20–30 cm. In the north of this island, ranges of the radioactivity of ^{137}Cs of the soils from four soil depths of the two red soils of Pinchen series and Longzhong series were $1.1 \pm 0.2 - 3.6 \pm 0.3$ and $5.9 \pm 0.2 - 7.4 \pm 0.2$ Bq kg^{-1}, respectively. Comparatively, in the center, that of the alluvial soil of Chunliao series the radioactivity was significantly lower ($0.75 \pm 0.02 - 1.2 \pm 0.1$ Bq kg^{-1}). This may be attributed to the higher clay and organic C contents and CEC values of the two red soils than that of the alluvial soil (Table 2). Additionally, higher amounts of fallouts from nuclear weapon tests in the north of this island of higher latitude (Lai *et al*., 1995) are also one of the reasons for higher radioactivity in the soils of north of this island.

In the east of this island, the arable soils are mainly developed from two kinds of parent materials, one is from the Central Mountains alluvial parent material and the other is from the Coastal Mountains alluvial parent material. Lin (1986) and Chen and Liu (1994)

Table 2 Physical and chemical properties of the soils

Sampling site	Soil series	Texture[a]	pH[b]	EC[c] (μS cm^{-1})	CEC[d] (cmol kg^{-1})	Org. C (g kg^{-1})	Exchangeable Ca (mg kg^{-1})	Exchangeable Mg (mg kg^{-1})	Exchangeable K (mg kg^{-1})	Exchangeable Na (mg kg^{-1})
Longtan, Taoyuan	Longzhong	SiC	4.71 c	396 a	9.5 b	17.7 a	350 c	120 b	150 a	745 a
Pingtung	Changhsing	LS	6.20 b	346 b	11.5 a	9.9 b	892 a	171 a	144 a	477 b
Puhsin, Taoyuan	Pinchen	C	4.59 c	107 d	7.6 c	7.4 c	282 d	47.4 d	112 b	334 c
Hsihu, Changhua	Chunliao	LS	7.01 a	269 c	4.4 d	5.9 c	783 b	71.6 c	50.8 c	169 d

The values are means and were examined on their differences at 5% significant level by least significant difference (LSD) test. Means followed by different lower case letters in a column indicate significantly different at 5% level.
[a]SiC: silty clay; LS: loamy sand; C: clay.
[b]pH value of the suspension with the ratio of soil to deionized water (w/v = 1:1).
[c]EC value of the filtrate obtained from the suspension measured for pH value.
[d]Longzhong and Pinchen soil series were measured the effective CEC.

Table 3 Citrate-bicarbonate-dithionite, oxalate, and pyrophosphate extractable Fe, Al, and Mn of the soils

Soil Series	Citrate-bicarbonate-dithionite extractable Fe (mg kg^{-1})	Citrate-bicarbonate-dithionite extractable Al (mg kg^{-1})	Citrate-bicarbonate-dithionite extractable Mn (mg kg^{-1})	Oxalate extractable Fe (mg kg^{-1})	Oxalate extractable Al (mg kg^{-1})	Oxalate extractable Mn (mg kg^{-1})	Pyrophosphate extractable Fe (mg kg^{-1})	Pyrophosphate extractable Al (mg kg^{-1})	Pyrophosphate extractable Mn (mg kg^{-1})
Longzhong	6881b	236 d	61.9 c	1638 a	1769 a	33.5 c	3175 a	2906 a	39.2 c
Changhsing	6391c	598 c	147 b	1486 b	420 c	104 b	284 d	140 c	65.7 a
Pinchen	22303a	3701 a	300 a	908 c	1383 b	96.0 b	2225 b	1911 b	46.1 bc
Chunliao	4801d	680 b	58.1 c	1503 b	261 d	140 a	661 c	103 c	54.3 b

The values are means and were examined on their differences at 5% significant level by least significant difference (LSD) test. Means followed by different lower case letters in a column indicate significantly different at 5% level.

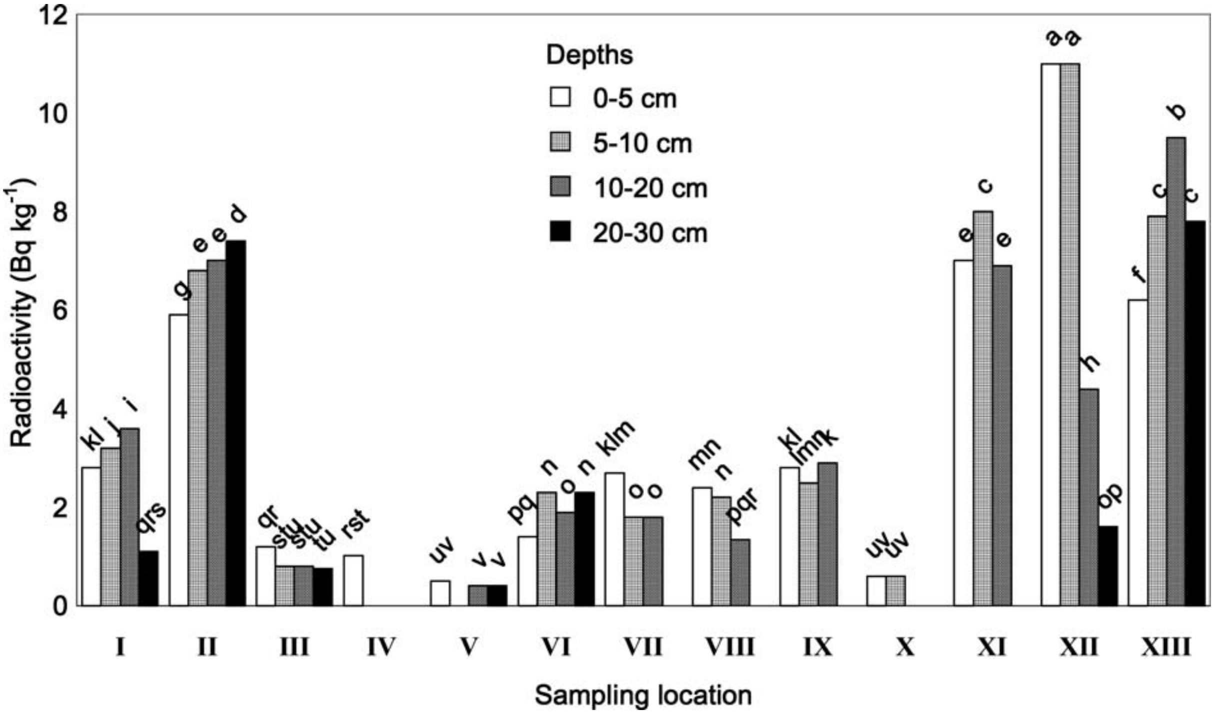

Fig. 2 Comparison of the radioactivity of ^{137}Cs in different profiles of some arable soils in Taiwan. Location I and II: Puhsin and Longtan Townships, respectively, Taoyuan County; III: Hsihu Township, Changhua County; IV, V, and VI: Kuanshan (Hungshih), Kuanshan (Tienkuang), and Chihshang Townships, respectively, Taitung County; VII, VIII, IX, and X: Yuli (Tayu), Yuli (Kuanin), Juisui (Wuho), and Juisui (Fuyuan) Townships, respectively, Hualien County; XI and XII: Sanhsing and Chuangwei Townships, respectively, Ilan County; XIII: Pinglin Township, Taipei County. The means of the radioactivity of ^{137} Cs in different profiles of the arable soils were examined on their differences at 5% significant level by least significant difference (LSD) test. Means with different lower case letters above the bars for the investigated depths between sampling sites are significantly different at 5% level.

reported the differences in fertility and characteristics of these two parent material-derived soils. In this study, although the soils collected in each township site were separately collected from two kinds of parent material-derived soils, the radioactivity of ^{137}Cs of all soils are significantly low (Taitung County, $0.4 \pm 0.1 - 2.3 \pm 0.1$ Bq kg^{-1}; Hualien County, $0.6 \pm 0.1 - 2.9 \pm 0.2$ Bq kg^{-1}) (Fig. 2). This indicates that the radioactivity of ^{137}Cs of the arable soils in Taitung and Hualien Counties are not correlated with fertility and characteristics of the parent material. Except for the soil of Fuyuan series, the radioactivity of ^{137}Cs of the soils from the profile depths of various soil series at Yuli and Juisui Townships, Hualiean County were significantly higher than those at Kuanshan Township, Taitung County (Fig. 2). This should be due to higher amounts of fallouts of higher latitude of this island (Lai *et al*., 1995).

In the north-eastern part of this island, ranges of the radioactivity of ^{137}Cs of the soils from the four profile depths of Sanhsing series (Sanhsing Townswhip, Ilan County), Chuangwei series (Chuangwei Township, Ilan county), and Tunglochuan series (Pinglin Township, Taipei County) were $6.9 \pm 0.1 - 8.0 \pm 0.3$, $1.6 \pm 0.1 - 11.0 \pm 0.2$, and $6.2 \pm 0.3 - 9.5 \pm 0.3$ Bq kg^{-1}, respectively (Fig. 2). The radioactivity of ^{137}Cs of the soil of Chuangwei series was the highest among the all collected arable soils in Taiwan and their differences were significant (Fig. 2). Furthermore, the highest radioactivity of ^{137}Cs of the soil of Chuangwei series was in the depths of 0–5 and 5–10 cm, which are the main depth ranges for nutrient absorption by crop roots. It thus merits close attention in investigating the transport of ^{137}Cs from the soil to edible part of the crops. The reason for the highest and higher radioactivity of ^{137}Cs in the soils from north-eastern part of this island could be due to the locations of Ilan County and part of Taipei County, which face against the northern and north-eastern seasonal winds annually from mid-autumn through mid-spring of next year

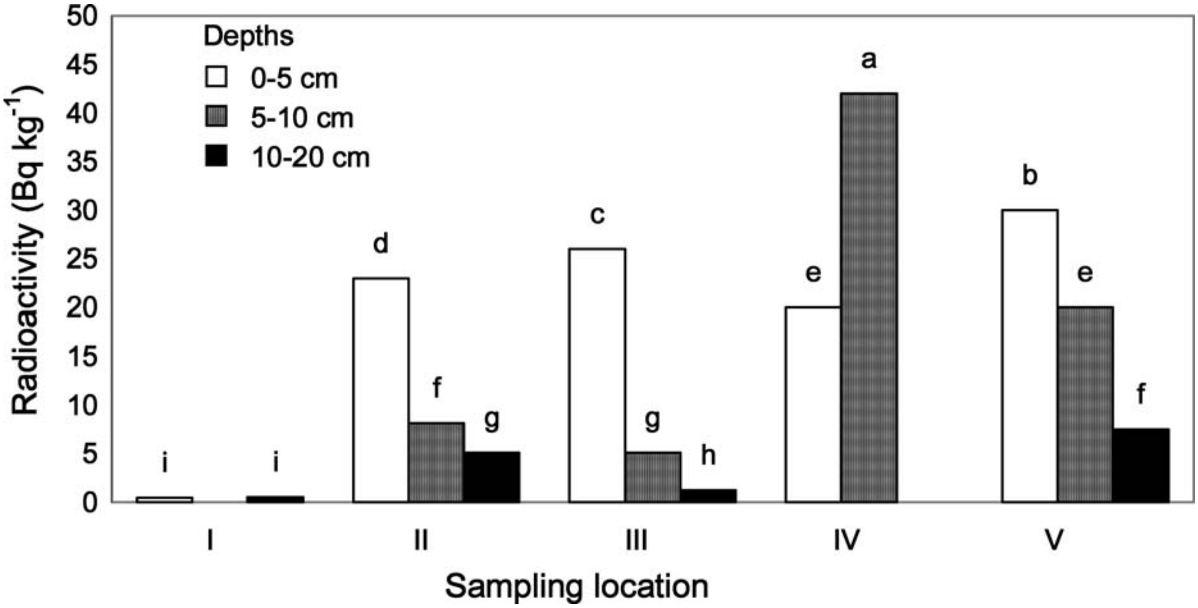

Fig. 3 Comparison of the radioactivity of ^{137}Cs in different profiles of two mountain forest soils in Taiwan. Location I: Wulin farm, Taichung County; II, III, IV, and V: at around the 1st, 2nd, 3rd, and 4th inflow mouths of the brooks, respectively, Yuanyang Lake Nature Preserve, among Ilan, Taoyuan, and Hsinchu Counties. The means of the radioactivity of ^{137}Cs in different profiles of the mountain soils were examined on their differences at 5% significant level by least significant difference (LSD) test. Means with different lower case letters above the bars for the investigated depths between sampling sites are significantly different at 5% level.

(Central Weather Bureau, 1988–2004). Moreover, except during unexpected typhoons with heavily storms, the yearly precipitations in the northern and north-eastern parts of Taiwan from mid-autumn through mid-spring of next year are much higher than those in the central, southern, and eastern parts of Taiwan (Central Weather Bureau, 1988–2004). Thus, more precipitation with strong northern and north-eastern seasonal winds annually from mid-autumn through mid-spring of next year may be partly contributing to the sedimentation of fallouts in Taipei and Ilan Counties. Furthermore, the north-eastern opened topography with the high Central Mountains at the back of Ilan and Taipei Counties (Fig. 1) suffers the seasonal winds with drizzle rain yearly, facilitating the sedimentation of fallouts in that area (Lu, 1994; Web page of Ilan County, 2003).

The radioactivity of ^{137}Cs of the forest soils from the profile depths of 0–5, 5–10, and 10–20 cm of the four sampling sites in the high mountains in Yuanyang Lake Nature Preserve, which is among the border of Ilan, Taoyuan, and Hsinchu Counties and also located in the north-eastern part of this island was in the range of $1.24 \pm 0.07 - 42 \pm 1$ Bq kg^{-1}, whereas that of the soils from the high mountains of Wulin Farm in the central part of this island was only of $0.48 \pm 0.08 - 0.53 \pm 0.07$ Bq kg^{-1} (Fig. 3). This clearly indicates the correlation of geographical location and topography of the sampling sites with the radioactivity of ^{137}Cs of the soils.

Lai *et al.* (1995) reported that amounts of fallouts from nuclear weapon tests in the north of this island of higher latitude were higher. Results of this study revealed hat the radioactivity of ^{137}Cs of arable soils and mountain soil in the northern and north-eastern parts of Taiwan were significantly higher than those of arable soils and mountain soil in other parts of this island (Table 1, Fig. 1, Fig. 2 and Fig. 3). The arable soils and mountain soil of the sampling sites in the northern and north-eastern parts of Taiwan are belonging to different soil groups in soil classification (Table 1). However, the radioactivity of ^{137}Cs in soils was significantly greater than those of the sampling sites in other parts of this island (Table 1, Fig. 1, Fig. 2 and Fig. 3). This indicates that radioactivity of ^{137}Cs in the investigated soils was not attributed to soil parent materials as well as cultivations of the soils. Thus, geographical location, topography, wind, and precipitation form, quantity, and

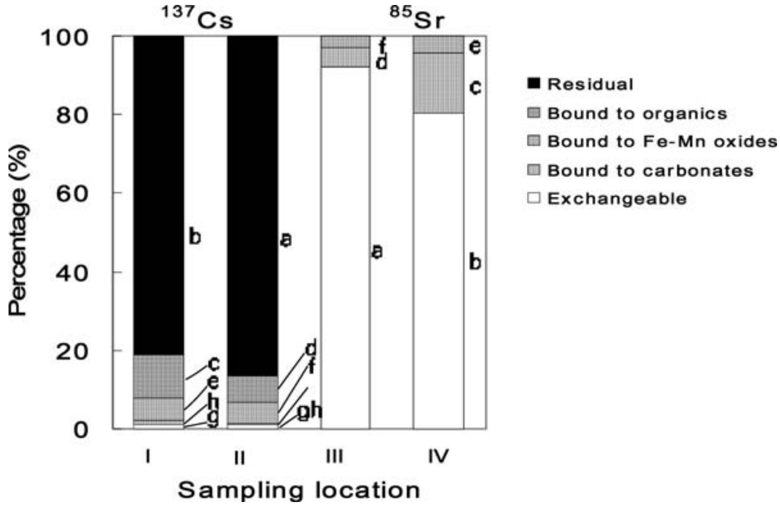

Fig. 4 The percentages of the solid-phase species of ^{137}Cs and ^{85}Sr in the soils. Location I: Longtan Township, Taoyuan County; II: Pingtung City; III: Puhsin Township, Taoyuan County; IV: Hsihu Township, Changhua County. The means of the percentages in different forms of solid-phase species of ^{137}Cs and ^{85}Sr in the soils were examined on their differences at 5% significant level by least significant difference (LSD) test. Means with different lower case letters beside the right-hand side of the column sections for the investigated soils are significantly different at 5% level.

duration play roles in governing the radioactivity of ^{137}Cs in soils of investigated areas.

3.3 Distribution of solid-phase species of ^{137}Cs or ^{85}Sr in the soils

A radionuclide undergoes the same reactions in soil as the non-radioactive isotope. Its adsorption depends on: (i) the properties of the ion in solution, and specifically whether it is negatively or positively charged or uncharged, and whether it forms complex ions in solution; (ii) the amounts of the oxides of iron, manganese, and aluminum and of the different clay aluminosilicates in soil; and (iii) the amount and nature of organic matter that is present (Wild, 1993; Schimmack *et al*., 2003). Mobile radioisotopes interact immediately with soil components while the others will reside in discrete fallout particles until they are released by weathering processes. There is much literature on studies relating to the uptake of mobile fallout cesium by clays (Poinssot *et al*., 1999). The solid-phase species of ^{137}Cs in its contaminated soil of Longzhong series (Longtan Township, Taoyuan County) and its amended soil of Changhsing series (Pingtung City) were mainly in the form of species bound to Fe-Mn oxides, bound to organics, and the residual forms and their differences were significant (Fig. 4). The percentages of the summed up radioactivity of these three species in the whole radioactivity of the soils of Longzhong series and Changhsing series were as high as 97.8 and 98.5%, respectively (Fig. 4). However, the solid-phase species of ^{85}Sr in its amended soils of Pinchen series (Puhsin Township, Taoyuan County) and of Chunliao series (Hsihu Township, Changhua County) were mainly in the species of exchangeable and minor amounts were bound to carbonates and to Fe-Mn oxides and their differences were significant (Fig. 4). The species of bound to organics and of the residual form were almost not detectable (Fig. 4). This great difference in the distribution of solid-phase species of ^{137}Cs contaminated or amended soil and of ^{85}Sr amended soil is mainly due to different reaction period of ^{137}Cs or ^{85}Sr with the soils. The ^{137}Cs contaminated or amended period was over three years, whereas the ^{85}Sr amended period was less than two months. The reaction of the contaminated or amended ^{137}Cs with the soil components increases the gradual formation of much more stable solid-phase species such as the forms bound to oxides and organics and the residual form, with increasing the reaction period. Comparatively, the reaction of the amended ^{85}Sr with the soil components was much shorter period, leading only to the formation of the unstable water-soluble and exchangeable forms. The radioactivity of the exchangeable species of ^{85}Sr in the whole radioactivity of ^{85}Sr of the soils of Pinchen series and Chunliao series were 92.1 and 80.5%, respectively

(Fig. 4). This significant difference in percentages is mainly attributed to higher clay and organic carbon contents of the former than the latter (Table 2). Due to calcareous characteristics and higher pH of the soil of Chunliao series, the percentage of the species of ^{85}Sr bound to carbonates was as high as 15.2% (Fig. 4). Furthermore, the percentage of the species of ^{85}Sr bound to oxides of the soil of Chunliao series was significantly higher than that of Pinchen series. This may be attributed to higher contents of oxalate-extractable Fe and Mn (noncrystalline forms of oxides) of the former than the latter (Table 3). The noncrystalline form of Fe and Mn oxides with substantial surface areas thus contribute to the reaction of soils of Chunliao series with ^{85}Sr.

4 Conclusions

The sequence of the radioactivity of ^{137}Cs in the profiles of arable soils in Taiwan was: Ilan and Taipei Counties in the north-eastern > Taoyaun County in the northern > Changhua County in the central, Tainan County in the southern, and Hualien and Taitung Counties in the eastern, and their differences were significant. The highest radioactivity of ^{137}Cs in the profiles of arable soil in Ilan County mainly correlates with its characteristic geographical location and topography of frequently facing northern and north-eastern seasonal winds with high precipitation annually from mid-autumn through mid-spring next year. Study on the radioactivity of ^{137}Cs of the forest soil from high mountains in the same area confirmed this correlation. Radioactivity of ^{137}Cs in the investigated soils is not attributed to soil parent materials as well as cultivations of the soils. However, geographical location, topography, wind, and precipitation form, quantity, and duration play roles in governing the radioactivity of ^{137}Cs in investigated soils. Textures and contents of organic matter and oxides of the arable soils significantly affect the radioactivity and distribution of ^{137}Cs in soil profiles. The reaction periods of ^{137}Cs and of ^{85}Sr with soil components influence the distribution of the solid-phase species of ^{137}Cs and ^{85}Sr in soil environment. Due to the similar chemical behavior of ^{85}Sr to that of ^{90}Sr, the data obtained in this study can be used to draw the conclusions of ^{90}Sr in the fallouts derived from nuclear weapon tests.

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