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Vertical distribution and migration of global fallout Pu in forest soils in southwestern China

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ABSTRACT

Soil samples collected in southwestern China were analyzed for Pu isotopes. The $^{240}Pu/^{239}Pu$ atom ratios were around 0.18, which indicated the dominant source of global fallout. Consistent sub-surface maximums followed by exponential decline of $^{239+240}Pu$ activities in the soil cores were observed. Most of the Pu has still remained in the 0–10 cm layers since its deposition. Convection velocities and dispersion coefficients for Pu migration in the soils were estimated by the convection–dispersion equation (CDE) model. The effective convection velocities and effective dispersion coefficients ranged from 0.05 to 0.11 cm/y and from 0.06 to 0.29 cm^2/y, respectively. Other factors that control the vertical migration of Pu in soil besides precipitation, soil particle size distribution and organic matter were suggested. Long-term migration behaviors of Pu in the soils were simulated. The results provide the Pu background baseline for further environmental monitoring and source identification of non-global fallout Pu inputs in the future.

1. Introduction

Plutonium isotopes are present in the environment as a result of nuclear weapon testing, nuclear fuel reprocessing (e.g., Mayak and Sellafield facilities) (Oughton et al., 2000; McCarthy and Nicholls, 1990) and nuclear facility accidents (e.g., the Chernobyl and Fukushima accidents) (Bunzl et al., 1995; Zheng et al., 2012). The nuclear weapon tests carried out in the early 1960s are the main source of Pu in the terrestrial environment and the total amount of $^{239+240}Pu$ released from the source in a global scale was estimated to be 11 PBq (UNSCEAR, 2000). Much attention has been paid to the evaluation of radiation risk of Pu in the environment (Yamamoto et al., 1999; Turner et al., 2003; Ketterer et al., 2004; Zheng et al., 2012) due to its strong radiological toxicity and long-term persistence (half-lives of ^{239}Pu and ^{240}Pu are 24100 y and 6561 y, respectively). Other studies have focused on its migration behavior in the environment (Bossew et al., 2004; Kaplan et al., 2004; Demirkanli et al., 2008; Ovsiannikova et al., 2010; Orzel and Komosa, 2014). The $^{240}Pu/^{239}Pu$ atom ratio varies significantly based on the source and production process, and thus it provides an important fingerprint for radioactive source identification (Kelley et al., 1999). For example, the $^{240}Pu/^{239}Pu$ atom ratio in weapon-grade Pu is typically below 0.05, while it is as high as 0.4 in mixed oxide fuels (Taylor et al., 2001). The global fallout Pu is characterized by a $^{240}Pu/^{239}Pu$ atom ratio of 0.18 (Kelley et al., 1999).

In the past few decades, the characterization of Pu isotopes in the terrestrial environment in many countries, especially in the areas around the former nuclear test sites and around the nuclear facilities, has been studied intensively (Yamamoto et al., 1999; Muramatsu et al., 2001; Turner et al., 2003). However, information about Pu distribution in the soils of China is limited. From 1964 to 1996, 45 nuclear tests, including 22 atmospheric nuclear tests were conducted at Nop Lor in northwestern China. These nuclear tests introduced test debris into the stratosphere as well as into local regions. Now there are 17 operating nuclear power reactors and 30 under construction in the coastal provinces of China. Construction of more reactors is planned in the inland provinces, such as Chongqing Municipality in southwestern China. As the Chernobyl nuclear accident caused long distance Pu contamination in the environment and the release of trace amount of Pu from the Fukushima nuclear accident was also observed in the areas around the nuclear power plant site, the distribution of Pu isotopes in the terrestrial environment of China needs to be investigated to provide baseline data.

Sha et al. (1991) measured total $^{239+240}Pu$ activities by alpha spectrometry in soils collected from several areas in eastern China.

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Zheng et al. (2009) reported the $^{239+240}$Pu activities and ^{240}Pu/^{239}Pu atom ratios in surface soils and a soil core collected in Gansu Province, in northwestern China and observed no significant influence of Chinese nuclear tests on the Pu contamination. More recently, Dong et al. (2010) and Xu et al. (2013) studied the vertical distribution of Pu isotopes in several soil cores collected from central China and northeastern China, respectively. To the best of our knowledge, to date, no study focusing on the distribution of Pu isotopes in soils from southwestern China has been conducted.

Understanding the vertical distribution and migration of anthropogenic radionuclides in soil is important for radiological assessment. Slow migration of anthropogenic radionuclides in soil gives rise to external dose for a long time, and results in the radionuclides being available to plant uptake. On the other hand, fast migration can lead to quick entrance of radionuclides into the underground water, thus increasing the potential risk of internal dose (Bossew and Kirchner, 2004). Since the early 1960s, migration models have been developed for predicting the migration of global fallout and regional fallout radionuclides in soil, of which the convection–dispersion equation (CDE) model has been most widely used. The CDE model takes account of the two mechanic processes of radionuclides in soil: convection and dispersion, which are related to the physical mechanics of radionuclide transport and the sorption of radionuclide in soil. The simulating parameters needed for this model for the radionuclides such as ^{134}Cs, ^{137}Cs, ^{90}Sr have been well addressed (Kirchner et al., 2009; Strebl et al., 2009; Szerbin et al., 1999; Schimmack and Marquez, 2006). However, the application of the CDE model for Pu migration prediction in soil is seriously constrained by the lack of field data (Strebl et al., 2009). In recent years, Pu has been considered as a new powerful tracer to replace ^{137}Cs in studies of desertification and soil erosion due to the easier Pu determination by today's highly sensitive mass spectrometers and the relatively long half-live of Pu (Hirose et al., 2003; Hoo et al., 2011; Xu et al., 2013). In order to further explore the feasibility of Pu as a tracer for environmental studies, the vertical migration behavior of Pu in soil needs to be better understood and the database for transfer parameters for Pu migration needs to be expanded.

In this work, we have studied the vertical distribution of Pu isotopes in soil core samples collected in forests in southwestern China in order to: (1) investigate the distribution characterization of Pu in forest soils; (2) establish the Pu background database in southwestern China for the possible radioactive source identification in the future; and (3) provide the migration parameters of Pu for the CDE model and predict the downward migration behavior of Pu in soil.

2. Materials and methods

2.1. Sample collection

Three soil core samples were collected from September 2011 to October 2011 in the forests at Guiyang (GY), Wulong (WL) and Zhongxian (ZX) sites from Guizhou Province and Chongqing Municipality in southwestern China. The sampling locations are shown in Fig. 1. The annual precipitations for these places ranged from 1100 to 1300 mm and the elevations were between 600 m and 1900 m above the sea. The GY and WL sites were in open areas in Changpoling National Forest Park and Xiannvshan National Forest Park, respectively, covered by native vegetation. In these two soil cores, plenty of vegetation roots were observed. The ZX site was situated on the top of a broad hill in the woodland, covered by pine needles. Small pine tree root branches were found in this soil core. All of the above sampling sites were without obvious human disturbances.

At each sampling site, one soil core was collected to a depth of 30 cm by a steel soil sampler ($d = 5$ cm). The top 0–10 cm depth of the soil cores was sliced at 2 cm intervals and 10–30 cm depth, at 5 cm intervals. For the two soil cores collected at the GY and WL sites, the top 10 cm soils mainly consisted of humus and the 10–30 cm depth soils were yellow/brown soils. The ZX soil core was very porous and the main component was sand. For each soil core, small portions from different layers were taken out and combined together for the analysis of particle size distribution with a laser particle size analyzer (Mastersize 2000, Malvern Instruments, UK) and pH values with a pH meter (D 14, Horiba Scientific, Japan). Detailed information about the sample sites and soil properties is summarized in Table 1.

2.2. Sample preparation and Pu measurement

The soil samples were firstly dried at 105 °C for 24 h. Then they were calcinated in a muffle furnace at 450 °C for 5 h to destroy the organic matter. The organic matter contents for the soil samples were determined by the mass losses before and after the ashing procedure. The chemical procedures for Pu separation in soil samples were based on our previous work (Bu et al., 2014). Briefly, a sample of about 2 g soil was weighed out and *ca.*1 pg ^{242}Pu was added as a yield monitor. Acid leaching (with 20 mL conc. HNO_3) was performed on a hot plate for at least 4 h at 160 °C. Then a two-stage anion-exchange chromatography method using AG 1X8 and AG MP-1 M resins was used for the separation of Pu from the sample matrix and the further purification of Pu. The final sample solution was dissolved in 0.8 mL 4% HNO_3, in preparation for Pu analysis. The overall chemical recoveries ranged from 48% to 86% with an average of 64% ± 9%.

We used the sector field (SF)-ICP-MS (Element 2, Thermo Finnigan, Bremen, Germany) in a low resolution mode ($m/\triangle m = 300$) for the analysis of Pu isotopes. The SF-ICP-MS was equipped with an APEX-Q high efficiency sample introduction system (Elemental Scientific Inc, Omaha, NE, USA) combined with a membrane desolvation unit (ACM) and a conical concentric nebulizer. The isotopes of interest (^{238}U, ^{239}Pu, ^{240}Pu, ^{242}Pu) were analyzed in the peak hopping mode and the peak tops of the masses were measured at 10% of their respective peak width. The detailed operational setup and parameters of this analytical system for Pu analysis have been given elsewhere (Zheng and Yamada, 2006). The sensitivity (^{238}U) for this system was about 1.2×10^7 cps ppb^{-1} and the detection limit for Pu determination was as low as 0.14 fg mL^{-1}. A Pu isotope standard solution (NBS-947) with a known ^{240}Pu/^{239}Pu atom ratio was used for mass bias correction. Two soil reference materials (IAEA-soil-6 and IAEA-375) were used for method validation.

2.3. The CDE migration model

The vertical migration of Pu with a concentration of C (Bq/cm^3) in soil can be characterized by a one-dimension convection–dispersion equation:

$$\frac{\partial C}{\partial t} = D\frac{\partial^2 C}{\partial x^2} - \upsilon\frac{\partial C}{\partial x} - \lambda C \quad (1)$$

where D (cm^2/y) is the effective dispersion coefficient; v (cm/y) is the effective convection velocity; λ is the decay constant; t (y) is the time since Pu deposition to the surface soil; x (cm) is the soil depth. The decay of Pu is very slow and the decay formula only affects the amount of Pu activity presented, not the migration parameters, so it can be ignored in the solution to Equation (1).

As the Pu isotopes in soil originated from human nuclear activities and the global fallout mainly occurred in the early 1960s,

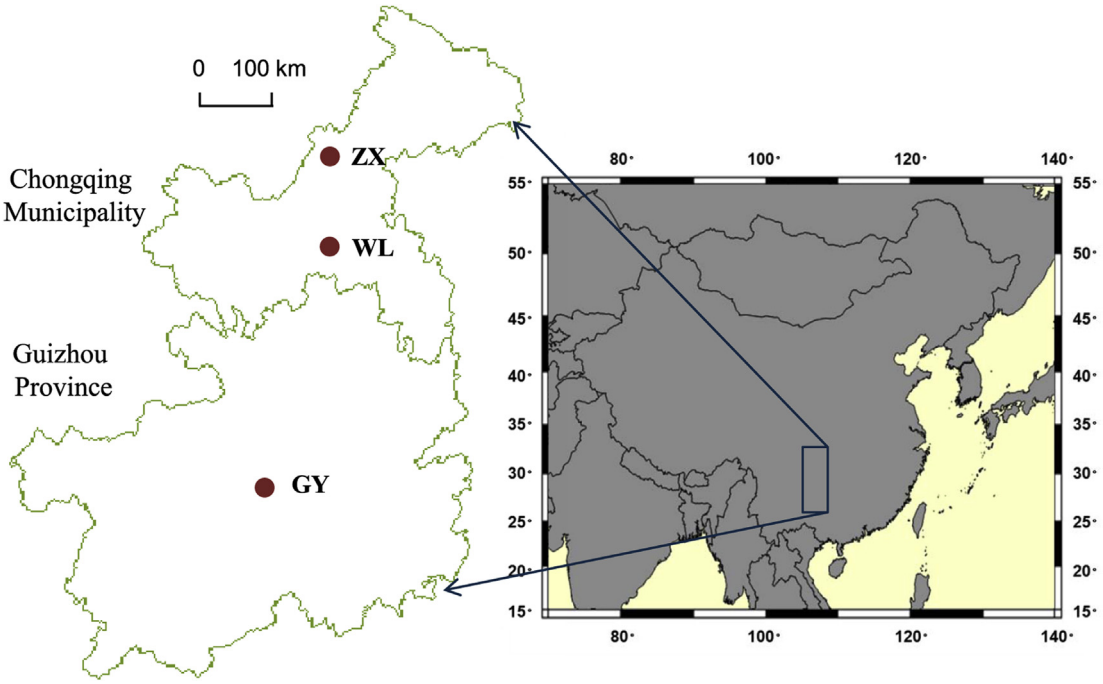

Fig. 1. Map showing the soil sampling sites in Guizhou Province and Chongqing Municipality in southwestern China.

the input of Pu to the soil can be treated like a single pulse. The following simplified solution has been widely used (Szerbin et al., 1999; Smith and Elder, 1999; Bossew and Kirchner, 2004):

$$C(x,t) = C_0 \frac{1}{2\sqrt{\pi D t}} e^{-\frac{(x-vt)^2}{4Dt}} \quad (2)$$

where C_0 is the amount of Pu present and it can be calculated by the measured Pu distribution in a soil core.

Based on the vertical distribution of Pu isotopes in a soil core, the effective dispersion coefficient and effective convection velocity can be fitted. When Equation (2) is used for modeling the migration of Pu in soil, important simplifications should be noted. (1) This model does not uniquely account for Pu sorption to soil but combines the retardation term in the dispersion (D) and convection (v) terms. As the Pu K_d values varies from different types of soil, the estimated parameters are highly specific to the sampling site. (2) The parameters D and v are assumed to be independent of time and soil depth. Although the conditions of soil, such as the organic matter content, the mineral composition may vary between different layers in a soil core, the estimated "mean" parameters over the considered soil depth, can still be typical for a soil/Pu system.

The measured $^{239+240}$Pu activities (mBq/g) were first converted to volumetric values (Bq/cm^3). The Pu inventories were calculated for the sampling depth (0–30 cm). Since the global deposition of $^{239+240}$Pu in the Northern Hemisphere peaked at the year 1963 (Hamilton et al., 1996), we chose 1963 to be the zero time and set $t = 48$ y. The three soil cores were fitted by using an unweighted nonlinear least-squares fitting procedure in commercial statistical software (Origin 8.6, OriginLab, USA).

3. Results and discussion

3.1. $^{239+240}$Pu activity and ^{240}Pu/^{239}Pu atom ratio

The analytical results of $^{239+240}$Pu activities, ^{240}Pu/^{239}Pu atom ratios and the organic matter contents in the soils collected in southwestern China are summarized in Table 2. The $^{239+240}$Pu activities in the surface (0–2 cm) soils collected from the three locations varied significantly. At the WL site, the surface $^{239+240}$Pu activity was 1.30 mBq/g, while at the ZX site, the surface $^{239+240}$Pu activity was much lower (0.171 mBq/g). In northeastern China, a wide range of $^{239+240}$Pu activities, from 0.027 to 0.938 mBq/g, have also been observed in the surface grassland (Xu et al., 2013). For all three sites, sub-surface (2–4 cm for WL and ZX sites and 4–6 cm for GY site) maximums of $^{239+240}$Pu activity were found followed by exponential decreases with depth. The concentrations of Pu below the depth of 20 cm became very low (<0.06 mBq/g).

The ^{240}Pu/^{239}Pu atom ratio is an important fingerprint for Pu source identification. For the northern hemisphere (0–30°N), the global fallout ^{240}Pu/^{239}Pu atom ratio was suggested to be 0.18 ± 0.02 (2σ) (Kelley et al., 1999). Unlike the variations of $^{239+240}$Pu activities, the ^{240}Pu/^{239}Pu atom ratios for all the soils from the top layer to the deeper layer at all three sites were relatively constant. The average ^{240}Pu/^{239}Pu atom ratios for the GY, WL and ZX sites were 0.190 ± 0.011 (1σ), 0.186 ± 0.004 (1σ) and 0.186 ± 0.012 (1σ), respectively, typically in the range of the global

Table 1
Information about the sampling sites and soil properties.

Sample site	Location	Sampling date	Elevation (m)	Precipitation (mm/y)	Clay (%)	Silt (%)	Sand (%)	pH
GY	26°39′23″N 106°40′10″E	14 Oct. 2011	1349	1130	13.8	81.2	4.9	4.77
WL	29°31′02″N 107°50′45″E	30 Sept. 2011	1852	1200	7.0	89.6	3.5	3.94
ZX	30°31′47″N 107°50′45″E	4 Oct. 2011	614	1214	4.4	31.1	64.6	3.95

Table 2
Organic matter, $^{239+240}$Pu activities and ^{240}Pu/^{239}Pu atom ratios in soil cores collected from southwestern China.

Sample site	Depth (cm)	Organic matter (%)	$^{239+240}$Pu activity (mBq/g)[a]	^{240}Pu/^{239}Pu atom ratio[a]	$^{239+240}$Pu inventory (Bq/m^2)
GY	0–2	18.6	0.316 ± 0.012	0.191 ± 0.007	63
	2–4	13.8	0.356 ± 0.011	0.182 ± 0.006	(0–30 cm)
	4–6	12.0	0.391 ± 0.017	0.194 ± 0.009	
	6–8	10.6	0.329 ± 0.016	0.188 ± 0.009	
	8–10	10.6	0.277 ± 0.014	0.188 ± 0.011	
	10–15	9.8	0.211 ± 0.011	0.175 ± 0.012	
	15–20	8.9	0.068 ± 0.005	0.183 ± 0.014	
	20–25	8.4	0.008 ± 0.003	0.202 ± 0.080	
	25–30	9.3	0.011 ± 0.002	0.209 ± 0.057	
WL	0–2	20.3	1.30 ± 0.05	0.181 ± 0.008	114
	2–4	17.0	1.44 ± 0.03	0.184 ± 0.005	(0–30 cm)
	4–6	15.0	0.667 ± 0.021	0.187 ± 0.006	
	6–8	14.5	0.286 ± 0.026	0.190 ± 0.010	
	8–10	13.5	0.094 ± 0.006	0.193 ± 0.016	
	10–15	12.8	0.069 ± 0.007	0.184 ± 0.020	
	15–20	11.7	0.065 ± 0.004	0.183 ± 0.014	
	20–25	12.4	0.058 ± 0.004	0.182 ± 0.012	
	25–30	11.9	0.045 ± 0.003	0.186 ± 0.011	
ZX	0–2	8.2	0.171 ± 0.008	0.185 ± 0.011	19
	2–4	4.9	0.185 ± 0.018	0.178 ± 0.025	(0–30 cm)
	4–6	4.5	0.139 ± 0.012	0.174 ± 0.015	
	6–8	3.8	0.039 ± 0.004	0.195 ± 0.022	
	8–10	4.0	0.029 ± 0.004	0.181 ± 0.028	
	10–15	4.0	0.032 ± 0.005	0.192 ± 0.032	
	15–20	3.8	0.016 ± 0.003	0.170 ± 0.044	
	20–25	3.4	0.005 ± 0.002	0.197 ± 0.088	
	25–30	3.8	0.006 ± 0.002	0.206 ± 0.080	

[a] Uncertainties represent 1σ error.

fallout value. Zheng et al. (2008) studied the vertical distribution of Pu isotopes in a sediment core in Lake Hongfeng (*ca.* 30 km from the GY site) in southwestern China and they found that the average ^{240}Pu/^{239}Pu atom ratio was 0.185 ± 0.009 (1σ). This result was consistent with the results we observed in the soils in southwestern China. In central and northeastern China, similar ^{240}Pu/^{239}Pu atom ratios have also been reported (Dong et al., 2010; Xu et al., 2013). The possible Pu contamination in China could be due to the global fallout and the regional fallout from the Lop Nor Chinese nuclear test site. Our ^{240}Pu/^{239}Pu atom ratio results in the soils suggested that the influence of the Chinese nuclear tests on the contamination of Pu in the investigated areas is very small, if any.

3.2. Depth profiles of $^{239+240}$Pu and inventories

The vertical distributions of Pu isotopes in the soil core samples were illustrated in Fig. 2. We can see that the distributions of Pu isotopes in all the soil cores showed the same trend: a subsurface maximum followed by exponential decrease. Most of the Pu (69%, 86% and 79% for the GY site, the WL site and the ZX site, respectively) in the investigated areas remained in the first 10 cm layers of the soil cores. It has been demonstrated that the organic matter is one of the important factors for the Pu sorption and retention in soil due to the complex formation of Pu with humic substances (Bunzl et al., 1995; Lee et al., 1996). As shown in Table 2, the organic matter in the top 10 cm layers of soil in the WL site ranged from 13.5% to 20.3%. The high organic matter could be the reason for the high proportion of global fallout Pu that was retained in the upper layer soils of the WL site. Lee et al. (1996) reported the vertical distribution of global fallout Pu in Korean soils and found that about 85% of the total Pu were in the first 10 cm layer. Dong et al. (2010) investigated Pu concentration in soil cores collected in Hubei Province in central China and they concluded that the proportion of Pu in the first 10 cm layer was more than 95% even more than 40 y after the global deposition. Our results were comparable with the results observed by Lee et al. (1996) and slightly lower than that found by Dong et al. (2010).

The inventories of $^{239+240}$Pu for the soil cores from the GY, WL and ZX sites were estimated to be 63 Bq m^{-2}, 114 Bq m^{-2} and 19 Bq m^{-2}, respectively. A compilation of anthropogenic radionuclide fallout data has been presented in the UNSCEAR (2000) report and the inventory of global fallout Pu within the 20–30°N band was suggested to be 36 Bq m^{-2}. Our results for the GY and WL sites were higher than the suggested global fallout value, while the result for the ZX site was almost half of it. However, the UNSCEAR suggested value is only an average of the observed results in the corresponding latitude band and should not be used as a model for a specific area (Bennett, 2002). The global fallout density of Pu at a given place could be due to the concentration in precipitation, the

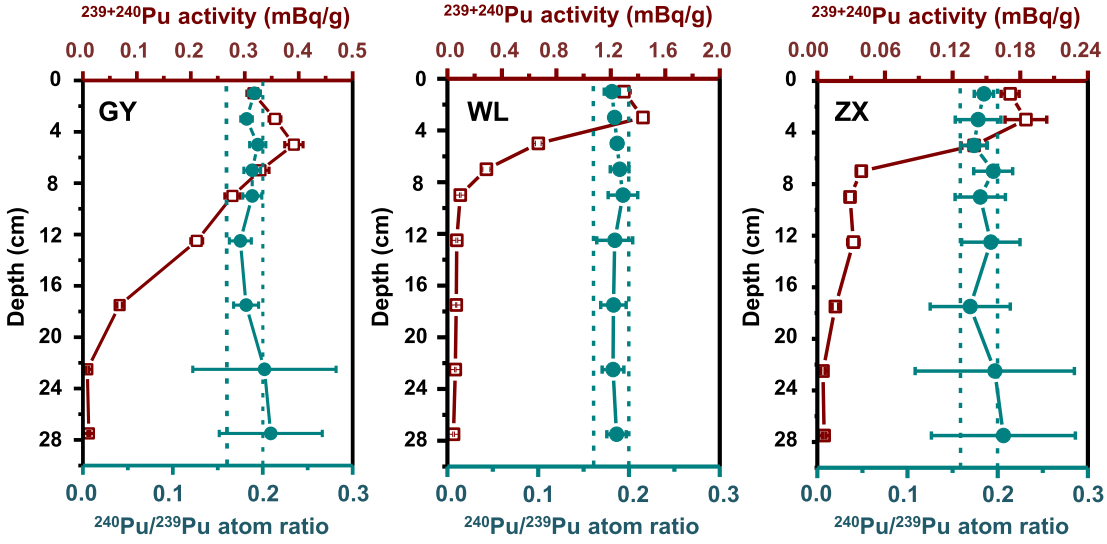

Fig. 2. Vertical profiles of $^{239+240}$Pu activities and ^{240}Pu/^{239}Pu atom ratios in the forest soil cores collected in the GY site in Guizhou Province and the WL and ZX sites in Chonqing Municipality in southwestern China. The dashed dark cyan lines indicate the global fallout ^{240}Pu/^{239}Pu atom ratio range of 0.18 ± 0.02. (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)

amount of precipitation in the interest time-interval, altitude, latitude etc. (Palsson et al., 2013). As the annual precipitations for the three sampling sites were similar, the high Pu inventories at the GY and WL sites could be due to the relatively high altitude. The ZX sampling site was situated on a broad hill and the main component of soil in this place is sand. The relatively low organic matter content in the ZX soil could be another reason for the lower retention of Pu at that place after the global fallout deposition.

3.3. Migration behaviors of Pu in the forest soils

3.3.1. Effective convection velocity and effective dispersion coefficient

As mentioned before, although the organic matter content varied slightly between different layers for the soil core samples (Table 2), the estimated "mean" parameters for the CDE model could still by typical for the specific sampling site. We fitted the CDE model with our measured vertical distributions of $^{239+240}$Pu activities in the soils. The fitting curves and the experiment results for Pu distribution in the soil cores are shown in Fig. 3. The goodness-fit-coefficients (R^2) for the GY, WL and ZX sites were 0.932, 0.984 and 0.950, respectively. The obtained convection velocities and dispersion coefficients with comparison to the literature values are shown in Table 3.

The effective convection velocities of global fallout Pu isotopes in the soils in southwestern China ranged from 0.05 to 0.11 cm/y and the effective dispersion coefficients ranged from 0.06 to 0.29 cm^2/y. Our results were comparable with the results (v, 0.15–0.20 cm/y; D, 0.02–0.11 cm^2/y) reported by Bossew et al. (2004) for Pu isotopes in soils of a grassland site in the Chernobyl exclusion zone.

Bossew and Kirchner (2004) analyzed more than 500 depth distributions of ^{137}Cs in soils of Austria and they found that the convection velocity and dispersion coefficient parameters resulting from the fitting of the CDE model could be grouped with regard to soils types and the parameter variability was considerably reduced within different groups. Kirchner et al. (2009) made a comprehensive review about the published data for the vertical distribution of ^{137}Cs in undisturbed grassland soils and calculated the migration parameters for different soil types by the CDE model. They found that the migration rate of ^{137}Cs in clay soils was lower than that of other soils. However, data about the migration ability of Pu isotopes in different types of soils are very limited. The particle size distribution and organic content for the soils collected from the GY site and WL site were similar (Tables 1 and 2). However, both the convection velocity and the dispersion coefficient of Pu at the GY site were larger than those at the WL site, suggesting the higher mobility of Pu at the GY site. It is interesting to find that although the particle size distribution and organic matter content for soils collected from the ZX site were much different from those of the WL site, there was no significant difference between their migration parameters. Our results suggested that there are other important factors besides particle size distribution and organic matter that control the migration ability of Pu in soils in our investigated areas. Those factors could include the chemical properties of the soil solution (pH, inorganic ion concentration), root water uptake and micro-organism and fungi bioturbation (Bunzl et al., 1995; Komosa, 1999; Demirkanli et al., 2008; Strebl et al., 2009; Matisoff et al., 2011). Further detailed studies are needed to specify the effect of these factors on the vertical migration of Pu in soil.

Table 3
The obtained parameters v and D for $^{239+240}$Pu in the forest soil cores fitted to the CDE model with the comparison to the literature values.

Sample site	Soil type	v (cm/y)	D (cm^2/y)	Reference
Chernobyl exclusion zone grassland	Unspecified	0.15–0.20	0.02–0.11	Bossew et al., 2004
GY forest	Silt	0.11	0.29	This study
WL forest	Silt	0.05	0.06	This study
ZX forest	Sand	0.05	0.09	This study

3.3.2. Downward migration prediction

Understanding and modeling the vertical downward migration of Pu in the investigated soils can help for the future monitoring of Pu in the environment of southwestern China and identifying the possible Pu released from the Chinese nuclear power reactors. We assumed that there would be no significant weather changes in the next 500 y in the sampling locations in this study and the soil hydraulic and Pu sorption properties in the deeper layers were similar to that of the top 30 cm layer. Note that the organic matter content in the soils below 30 cm may be lower than that of the upper layers, thus the predicted migration behaviors of Pu in the soil cores could be slightly underestimated. Based on the CDE model and the estimated migration parameters obtained from our experiment results, we simulated the downward migration of Pu in the soils from the three sampling sites at different times (50 y, 100 y, 200 y, 500 y after the global fallout deposition). The simulated results are illustrated in Fig. 4.

We can see that the Pu isotopes migrate downwards slowly in soil with passing time. For all three studied sites, after 50 y the

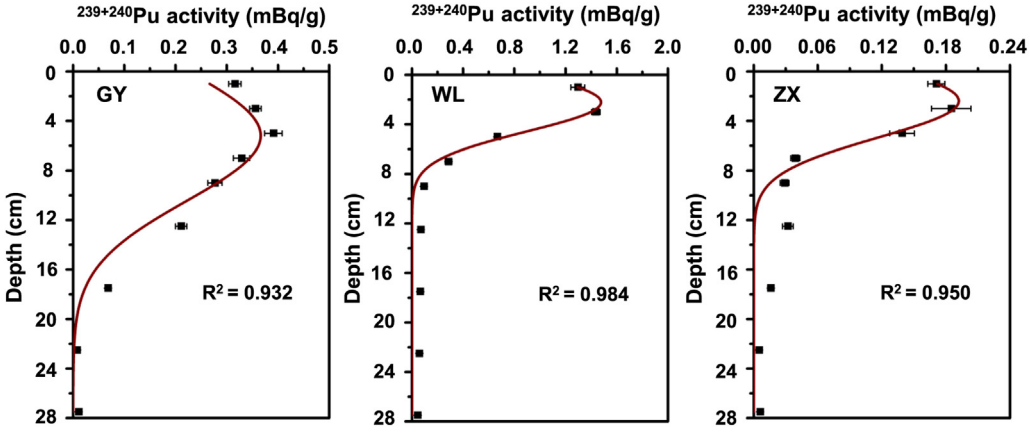

Fig. 3. Vertical distribution of $^{239+240}$Pu activities in the three soil cores and the CDE model fitted curves.

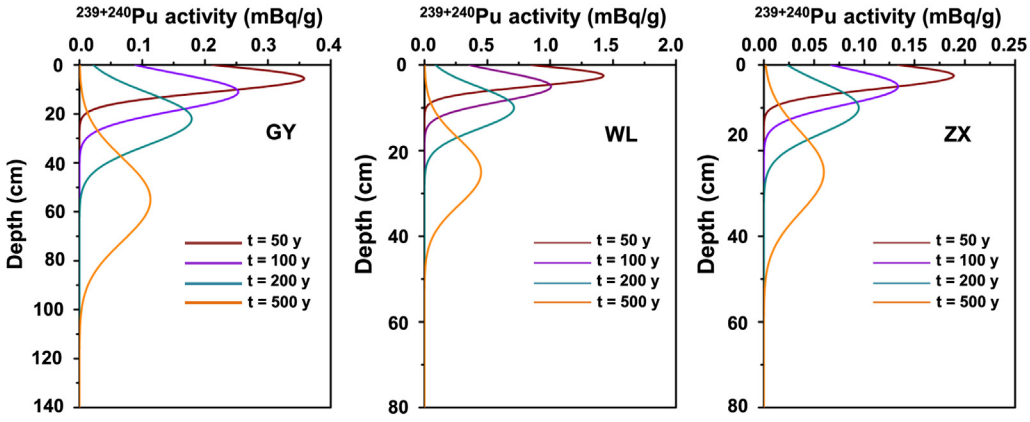

Fig. 4. Illustration of the downward migration behaviors of Pu in forest soils by the CDE model after deposition in 1963.

surface $^{239+240}$Pu activities would decrease to become *ca.* half of their current values. Most of the Pu would stay in the upper 20 cm layer soils at the WL and ZX sites during the next 150 y and the concentration peak of Pu would cross the 20 cm depth 500 y after the global fallout deposition. The downward migration of Pu in the soil at the GY site was faster than that of the other two sites. The concentration peak of Pu would cross the 20 cm depth 200 y after the deposition and would reach the 55 cm depth another 300 y later. These predicted results can provide background information for Pu distribution in soils in southwestern China and be used for source identification of the non-global fallout Pu input in the environment.

4. Conclusions

In this study, for the first time, we investigated the vertical distribution of $^{239+240}$Pu activities and ^{240}Pu/^{239}Pu atom ratios in the soil cores collected in the forests in Chonqing Municipality and Guizhou Province in southwestern China. The ^{240}Pu/^{239}Pu atom ratios in the soil cores indicated that the global fallout was the source for Pu contamination in these areas. Based on the CDE model and the measured results, we calculated the migration parameters of Pu in these forest soils and then we simulated the long term downward migration behavior of Pu. The convection velocities and dispersion coefficients obtained in our study expand the application of the CDE model for the prediction of Pu migration in soil. The experiment data set and the simulated results for Pu distribution in soils provided the background baseline for further environmental monitoring and the evaluation of the source term of possible non-global fallout Pu inputs in the future.

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