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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,