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Distinctive distributions and migrations of $^{239+240}$Pu and ^{241}Am in Chinese forest, grassland and desert soils

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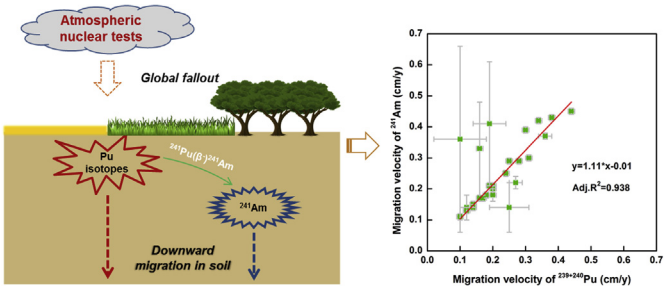
HIGHLIGHTS

- Distributions of Pu and Am in diverse types of Chinese soils were investigated.
- Higher apparent dispersion coefficients were observed for soils with lower pH.
- Mean migration velocities of Pu and Am were similar, being less than 0.3 cm/y.
- Migration results were compatible with short-term simulation studies in China.

GRAPHICAL ABSTRACT



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ABSTRACT

The vertical distributions and downward migrations of the global fallout derived $^{239+240}$Pu and ^{241}Am in diverse types of Chinese soils (forest, grassland and desert) were studied. The mean $^{239+240}$Pu and ^{241}Am activity concentrations in the investigated soil cores were 0.28–0.69 mBq/g and 0.13–0.37 mBq/g, respectively, while the accumulative inventories were 61.53–138.99 Bq/m^2 for $^{239+240}$Pu and 28.29 –61.05 Bq/m^2 for ^{241}Am. The convection-dispersion equation (CDE) was used to calculate the migration parameters of $^{239+240}$Pu and higher apparent dispersion coefficients (D) were observed for the acidic forest soils compared with the alkaline grassland and desert soils; meanwhile a compartment model was employed to compare the migration of $^{239+240}$Pu and ^{241}Am in successive soil layers which showed that the migration behaviors of $^{239+240}$Pu and ^{241}Am were rather similar; both velocities were less than 0.3 cm/y in diverse types of soils and these findings were compatible with those of short-term laboratory simulation experiments in China.

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1. Introduction

Plutonium (Pu) has been released to the environment worldwide mainly as a result of nuclear weapon tests in the last century, by which the total amount of $^{239+240}$Pu was approximately 11 PBq

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(UNSCEAR, 2000). Unlike the fallout $^{239+240}$Pu that was directly deposited to the earth's surface during the atmospheric nuclear weapon tests, ^{241}Am ($T_{1/2} = 432.2$ y) (Hou and Roos, 2008) has been steadily increasing in the environment as a consequence of the beta-decay of ^{241}Pu ($T_{1/2} = 14.4$ y) (Hou and Roos, 2008) produced concurrently with other Pu isotopes during the nuclear explosions. The produced ^{241}Am content in the environment is expected to reach its maximum in the year 2042 (Zheng et al., 2012). On the one hand, considering their long half-lives and high radiotoxicities, the levels of $^{239+240}$Pu and ^{241}Am in the environment are of scientific concern regarding their long-term radiation risk assessments; on the other hand, since the ^{240}Pu/^{239}Pu atom ratio and ^{241}Am/$^{239+240}$Pu activity ratio vary from different sources, information on these characteristic ratios could be regarded as radioactive fingerprints to distinguish possible contamination once a nuclear accident happens (Zheng et al., 2012). Therefore, it is meaningful to enrich the database of $^{239+240}$Pu and ^{241}Am distributions in the diverse natural environments in China. Although many data have been reported on the distributions of $^{239+240}$Pu in several parts of China (Bu et al., 2014, 2015; Dong et al., 2010; Sha et al., 1991a, 1991b; Xu et al., 2013, 2017; Zheng et al., 2009), studies on ^{241}Am are very scarce. To our knowledge, only Sha et al. (1991a; 1991b) have reported the activity concentrations of $^{239+240}$Pu and ^{241}Am in soils collected in 1990 in northern and eastern China. Further investigations are necessary to renew the knowledge about the current status of ^{241}Am in Chinese environments.

Soil is a principal reservoir of radionuclides present in terrestrial environments. Information on their vertical migration in soil is valuable since this process controls the long-term behaviors of these radionuclides in ecosystems. Furthermore, comprehensive understanding of the environmental behaviors of long-lived radionuclides (such as ^{239}Pu, ^{240}Pu and ^{241}Am) is crucial for the management of highly radioactive waste disposal. Actually, simulation experiments under controlled field or laboratory conditions with artificially added Pu and Am tracers have been conducted in China to acquire valuable information on their mobility under specific scenarios (Guo et al., 2003; Li et al., 2004a, 2004b). Nevertheless, in spite of the advantages of the controlled experimental conditions, it is very difficult to ensure realistic environmental conditions in this way, especially for long-term investigations. For this reason, it is useful to study the behaviors of radionuclides that have been introduced into an actual environment for many years. Benefitting from having the same source of radionuclides, similar deposition density on regional scales and long migration times (decades) since their initial deposition onto the soil, the global-fallout-derived radionuclides are appealing tools for worldwide comparative studies on their migration behaviors in different environments and ecosystems.

Principally, the basic physical processes behind the migration of radionuclides in soil include convection through flowing pore water, dispersion caused by spatial variations of convection, diffusion of radionuclides within the fluid and physical-chemical interaction with soil particles (Kirchner et al., 2009). There are two models that have been widely used for modeling the migration of radionuclides, i.e. the convection-dispersion equation (CDE) and the compartment model. The CDE is established based on the actual physical processes of the radionuclides in the soil cores and has analytical solutions for specific initial and boundary conditions (Bossew and Kirchner, 2004). It has been widely employed to obtain the migration parameters of ^{137}Cs and ^{90}Sr which have been compiled by the IAEA in the technical report series IAEA TRS-472 (IAEA, 2010). However, the application of CDE for Pu migration is seriously limited by the lack of field data (Strebl et al., 2009). The application of CDE for ^{241}Am is much more complicated than that for Pu because not only the migration of ^{241}Am is involved, but also its ingrowth from ^{241}Pu should be considered. As a way out, the migration of ^{241}Am could be roughly evaluated by the compartment model without taking into account detailed migration mechanisms. The compartment model is a black-box approach described by a series of linear first-order differential equations with which the residence times of the radionuclides in each soil layer can be calculated (Boone et al., 1985; Kirchner, 1998). To date, reports on the migration of Pu isotopes and ^{241}Am in Chinese field environments are rather limited. To our knowledge, only Bu et al. (2014) have calculated the migration parameters of Pu in forest soils in southwestern China with CDE, while no results of ^{241}Am migration in undisturbed Chinese soils have been reported yet. On account of the large diversities of Chinese environment, it is informative to compare the migration parameters of Pu and Am in different environments to check whether the environmental difference could lead to distinct migration behaviors of Pu and Am.

In this work, we investigated the vertical distributions and downward migrations of $^{239+240}$Pu and ^{241}Am in four areas in China. These areas were chosen for their large diversities in environmental condition such as precipitation, soil pH, plantation coverage etc. Sources of $^{239+240}$Pu and ^{241}Am in these areas were identified. The migration of Pu was quantified by the physically based CDE. Besides, for the first time, the migration behaviors of $^{239+240}$Pu and ^{241}Am in Chinese soils in diverse environments (forests, grassland, and desert) were compared with the compartment model.

2. Materials and methods

2.1. Sample collection

Four soil cores were collected from diverse environments (two forests, one grassland and one desert) in different areas of China more than 1000 km from each other. The two core samples from Guiyang (GY) city (106°40′1″E, 26°39′23″N) and Guazhou (GZ) city (95°44′57″E, 40tyincepl) were collected in the previous work of Bu et al. (2014, 2015) in 2011 while the other two were sampled in 2014 (QS) from Qinshan (120°56′42″E, 30°26′17″N) and in 2015 from Chengde (CD) city (117°13′52″E, 42°24′13″N), respectively. The sample information and locations are presented in Table 1 and Fig. A.1. For GY, GZ and QS cores, the depth interval was 2 cm for the initial 10 cm and 5 cm for the subjacent soils. For the CD sample that was collected in another sampling campaign, the whole soil core was divided into 2 cm intervals. Before further analysis, soil samples were dried and passed through 2 mm sieve to remove gravels and plant roots.

2.2. Chemical separation and analysis of Pu isotopes and ^{241}Am

Pretreated soil samples were firstly ashed in an oven at 450 °C for 4 h to decompose organic matter (Wang et al., 2015). A HNO_3 leaching method was used to release Pu and Am from soil samples. The Pu and Am analysis procedures were described in Appendix A and more details can be referred to our previous work (Wang et al., 2016, 2017). Both Pu isotopes and Am fractions were analyzed by SF-ICP-MS. The IAEA-soil-6 reference material was used for quality control of the analytical methods and results were presented in Table A.1.

2.3. Model fitting

The migrations of Pu isotopes in these soil cores were quantified by two models, viz. the CDE and the compartment model. The CDE

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