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Plutonium concentration and $^{240}Pu/^{239}Pu$ isotopic ratio in the surface soils from the Jiuquan region in northwestern China

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Abstract Surface soil samples collected in the Jiuquan region in the downwind area of the Chinese nuclear test site (CNTs) were analyzed for Pu isotopes. The $^{239+240}Pu$ activities ranged from 0.025 ± 0.009 to 0.89 ± 0.16 mBq g^{-1}, varying significantly with different sampling sites. The Dunhuang city that is located in the southwestern part of the Jiuquan region received the heaviest Pu deposition ($^{239+240}Pu$ activities, 0.23–0.89 mBq g^{-1}). Most of the $^{240}Pu/^{239}Pu$ isotopic ratios were similar with that of the global fallout. However, the low values (0.080–0.147) observed in three sampling sites further supported the finding of Pu originated from CNTs in that region.

Keywords $^{239+240}Pu$ activity · $^{240}Pu/^{239}Pu$ isotopic ratio · Surface soil · Northwestern China · Chinese nuclear tests

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Introduction

Anthropogenic Pu isotopes are present in the environment mainly as a result of nuclear weapon tests, nuclear power plant accidents and nuclear fuel reprocessing [1–3]. The main source for Pu contamination is the nuclear weapon tests carried out in the early 1960s and the total amount of $^{239+240}Pu$ released from this source has been estimated to be 15 PBq [4]. Pu isotopes are characterized by radiological significance and high chemical toxicity, thus the distribution and characterization of the nuclear test released Pu isotopes in the environment attracted extensive scientific and public attentions [5–8].

From 1964 to 1980, 22 atmospheric tests have been conducted by China in the Lop Nor Chinese nuclear test site (CNTs) [4]. Radioactive clouds formed in the CNTs moved to the east passing through the Jiuquan region in Gansu province in northwestern China [9], causing extra regional fallout in this region. A large scale radiation investigation focused on the distribution of radionuclides in soil samples was carried out in the Jiuquan region to assess the cumulative effective dose from the CNTs 20 years ago after that China joined the PTBT (Partial Test Ban Treaty) signatory nuclear nations in 1980. The results revealed that *ca.* 70 % of ^{137}Cs in this region originated from the CNTs [9]. However, due to the limitation of measurement techniques, Pu isotopes were not included in this investigation. A pilot study of $^{239+240}Pu$ activities in soils in Xinjiang province found a high $^{239+240}Pu$ inventory in a soil core collected in the downwind area of the CNTs and the attribution of Pu from the CNTs was suggested [10].

In recent years, the development of mass spectrometry makes it possible to accurately measure not only the $^{239+240}Pu$ activity but also the $^{240}Pu/^{239}Pu$ isotopic ratio. The $^{240}Pu/^{239}Pu$ isotopic ratio varies significantly based on

the source and production process, thus serves as an important fingerprint for Pu source identification. For instance, the $^{240}Pu/^{239}Pu$ isotopic ratios from nuclear weapon-grade materials were normally below 0.07 [11]; the $^{240}Pu/^{239}Pu$ isotopic ratios from the Nevada test site, Semipalatinsk test site and the Pacific proving grounds in the North Pacific were reported to be 0.054–0.063, 0.025–0.072 and 0.30–0.36, respectively [5, 6, 12]. The global fallout Pu is characterized by a $^{240}Pu/^{239}Pu$ isotopic ratio of 0.18 [13].

Several studies have been conducted for the distributions of Pu isotopes in the soils from central China, northeastern China and southwestern China, which are far away from the CNTs. The obtained $^{240}Pu/^{239}Pu$ isotopic ratios were around 0.18, suggesting that Pu isotopes from these areas originated from global fallout [14–16]. For northwestern China, Jin et al. [17] firstly observed a low $^{240}Pu/^{239}Pu$ isotopic ratio of 0.16 in a surface soil collected from Gansu province. However, in the soils collected from Lanzhou (in downwind areas *ca.* 1200 km from the CNTs) in Gansu province and from Xinjiang province (in upwind areas 200–400 km from the CNTs), the $^{240}Pu/^{239}Pu$ isotopic ratios were found to be consistent with that of global fallout, revealing that the contribution of the CNTs released Pu to these areas was negligible [18, 19].

Our previous work focused on the vertical distributions of $^{239+240}Pu$ activities and $^{240}Pu/^{239}Pu$ isotopic ratios in the soil cores collected in the Jiuquan region in Gansu province, which is the nearest region with human habitation downwind from the CNTs [8]. The results demonstrated that this area received regional fallout Pu from the CNTs and the $^{240}Pu/^{239}Pu$ isotopic ratios from the CNTs were slightly lower than that of global fallout. In this study, we report the distribution of Pu isotopes in the surface soils collected in the Jiuquan region in order to establish a more comprehensive database for Pu distribution in the Jiuquan region and further understand the Pu contamination from the CNTs.

Experimental

The Jiuquan region is dominated by a typical continental arid climate and the annual precipitation is below 160 mm [20]. In July 2011, surface soil samples (0–2 cm) were collected by a steel soil sampler ($d = 5$ cm) at eight sampling sites (JQ, 39°45′45″N 98°25′27″E; JYG, 39°47′34″N 98°08′25″E; QQ, 39°47′51″N 98°07′45″E; GZ1, 40°32′43″N 95°48′50″E; GZ3, 40°24′33″N 95°44′57″E; GZ4, 40°34′48″N 95°45′00″E; DH1,40°09′48″N 94°53′24″E; DH2, 40°03′44″N 94°47′33″E) in the Jiuquan region, *ca.* 300–500 km east of Lop Nor (Fig. 1). All the sampling locations were in open areas covered by sparse vegetation (mainly grass and sand jujube trees). One surface soil sample was collected at JYG and DH1 sampling sites, respectively. For the other sampling sites, two to four surface soil samples were collected at each site with a distance of *ca.* 50 m from each other. As part of our project, soil core samples were collected in this region as well [8].

The soil samples were firstly dried at 105 °C for 24 h and then passed through a 20 mesh sieve to remove coarse materials. Then they were calcinated in a muffle furnace at 450 °C for 5 h to destroy the organic matter. The soil sample preparation procedures were revised based on our previous work [22] (Fig. 2). Briefly, about 1–3 g soil was weighed out and *ca.*0.57 pg ^{242}Pu was added as a yield tracer. Acid digestion (with 20 mL conc. HNO_3-1 M HF) was performed on a hot plate at 160 °C for at least 4 h. Then the sample was dissolved with 8 M HNO_3 and Pu was converted to the tetravalent state with $NaNO_2$. After that, boric acid was added and the sample solution was heated to avoid the complex of Pu fluoride. A two-stage anion-exchange chromatography method using AG 1X8 and AG MP-1 M resins was employed for the separation of Pu from the sample matrix and the further purification of Pu. The final sample solution was prepared in 0.8 mL 4 % HNO_3. The overall chemical recovery for the sample preparation procedures was *ca.* 60 %.

A sector field (SF)-ICP-MS (Element 2, Thermo Finnigan, Bremen, Germany) was used in a low resolution mode ($m/\triangle m = 300$) for the analysis of Pu isotopes. In order to improve the sensitivity of the measurements, a high efficiency sample introduction system (APEX-Q, Elemental Scientific Inc., Omaha, NE, USA) combined with a conical concentric nebulizer was used. The isotopes of interset (^{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. More detailed operational setup and parameters of the sample introduction system and the SF-ICP-MS for Pu analysis have been given previously [23]. A Pu isotope standard solution (NBS-947) with a known $^{240}Pu/^{239}Pu$ isotopic ratio of 0.242 was used for mass bias correction and two soil reference materials (IAEA-soil-6 and IAEA-375) were used for method validation. The analytical results of Pu isotopes in the soil reference materials by our method are shown in Table 1 together with the certified and information values. We can see that both the $^{239+240}Pu$ activities and the $^{240}Pu/^{239}Pu$ isotopic ratios agreed well with the certified and the literature values.

Results and discussion

Distribution of $^{239+240}Pu$ activities and $^{240}Pu/^{239}Pu$ isotopic ratios in the surface soils

The analytical results of $^{239+240}Pu$ activities and $^{240}Pu/^{239}Pu$ isotopic ratios in the surface soils collected in the Jiuquan

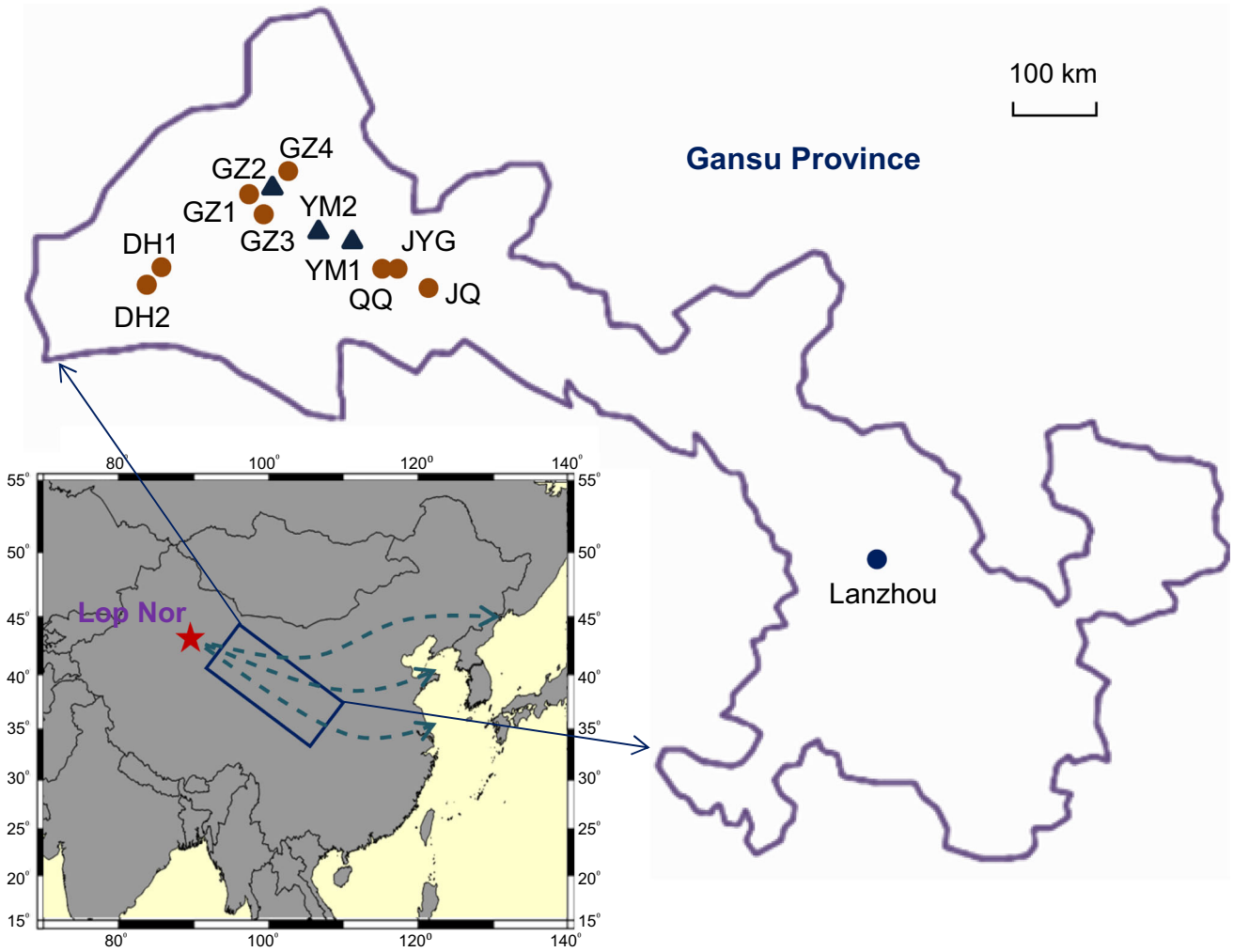

Fig. 1 Map showing the surface soil sampling sites in the Jiuquan region of Gansu province in northwestern China. The *dashed line* represents the typical pathway of the radioactive clouds after the Chinese nuclear tests [21]; The *triangle points* redrawn from [8] represent the soil core sampling sites; Lanzhou site is cited from [19]

region in northwestern China are summarized in Table 2 and shown in Fig. 3. The $^{239+240}$Pu activities ranged from 0.025 ± 0.009 to 0.89 ± 0.16 mBq g^{-1} with an average of 0.37 ± 0.32 mBq g^{-1} (1σ). Xu et al. [15] studied the distribution of Pu isotopes in the surface grassland soils collected from the Liaodong Bay in northeastern China. The $^{239+240}$Pu activities ranged from 0.023 to 0.938 mBq g^{-1}. Yang et al. [28] investigated the concentration of Pu in the surface soils from central-eastern Japan and found that the $^{239+240}$Pu activities were 0.004–1.46 mBq g^{-1}. The $^{239+240}$Pu activity results observed in the Jiuquan region in northwestern China were comparable with that in northeastern China and central-eastern Japan. For the soils (0–10 cm) collected 100–300 km away from the Semipalatinsk test site, the $^{239+240}$Pu activities were reported to be 1–4 mBq g^{-1}, much higher than that from the Jiuquan region due to the more intensive nuclear tests conducted at the Semipalatinsk test site [5]. Most of the ^{240}Pu/^{239}Pu isotopic ratios ranged from 0.165 ± 0.012 to 0.193 ± 0.053 with a mean of 0.178 ± 0.009 (1σ), similar to that of the global fallout and the results observed in the atmospheric air over Switzerland collected in recent years (0.18) [29]. However, in three sampling sites, low ^{240}Pu/^{239}Pu isotopic ratios (0.147 ± 0.009, 0.080 ± 0.007 and 0.132 ± 0.044 for GZ1-1, GZ4-2 and GZ4-3, respectively) were observed. These values were obviously lower than that of global fallout and the results observed in other areas of China [13–16].

The $^{239+240}$Pu activities from different sampling sites in the Jiuquan region varied significantly. At DH2 sampling site, the $^{239+240}$Pu activities exceeded 0.8 mBq g^{-1} while at the site of JQ, the $^{239+240}$Pu activities were typically below 0.05 mBq g^{-1}. In our previous study, we investigated the vertical distributions of Pu in the soil core samples collected in the Jiuquan region [8]. For the two soil

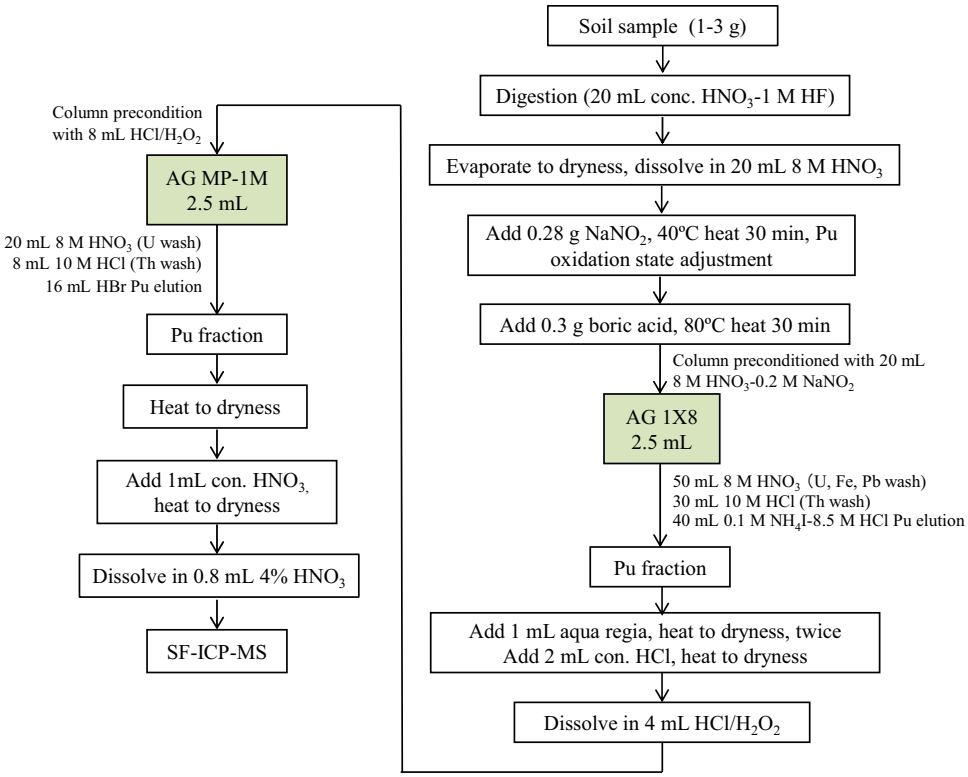

Fig. 2 Flow chart of the analytical procedure for the determination of Pu in soil samples

Table 1 Results of the two soil reference materials measured by our method and the certified and information values

Sample	$^{239+240}$Pu activity (mBq g^{-1})[a]	^{240}Pu/^{239}Pu atom ratio[a]
IAEA-soil-6-1	1.02 ± 0.07	0.191 ± 0.017
IAEA-soil-6-2	0.99 ± 0.04	0.181 ± 0.011
IAEA-soil-6-3	1.01 ± 0.05	0.191 ± 0.014
IAEA-soil-6-4	1.02 ± 0.04	0.190 ± 0.013
IAEA-375-1	0.27 ± 0.01	0.292 ± 0.017
IAEA-375-2	0.34 ± 0.01	0.268 ± 0.020
Certified and/or information values		
IAEA-soil-6	0.96–1.11	0.186–0.212[b]
IAEA-375	0.26–0.34	0.22–0.31[c]

[a] Uncertainties represent 1σ error; [b] Data cited from [24, 25]; [c] Data cited from [26, 27]

cores collected at DH2, the $^{239+240}$Pu activities in the surface soils (0–2 cm) were 0.833 ± 0.022 and 1.47 ± 0.06 mBq g^{-1}, respectively, which were consistent with the results observed in this study. Two extremely high Pu inventories (485 and 546 Bq m^{-2} for 0–30 cm), almost ten times of the global fallout value, were also observed at this site [8]. Considering the arid climate and low precipitation in the Jiuquan region, these results suggested that this area received extra Pu contamination besides global fallout. Although the $^{239+240}$Pu activities from different sampling sites varied, the results for the surface soil samples collected in a same site were similar. For example, the $^{239+240}$Pu activities for the three samples DH2-S1, DH2-S2 and DH2-S3 collected in the Dunhuang city were 0.87 ± 0.02, 0.84 ± 0.10 and 0.89 ± 0.16 mBq g^{-1}, respectively. These results demonstrated that the Pu analytical results obtained from the collected samples in this study could represent the real level of Pu in the surface soils of the sampling sites although the sample amount was relatively small.

As shown in Fig. 3, most of the ^{240}Pu/^{239}Pu isotopic ratios in the surface soils collected in the Jiuquan region were similar to that of global fallout. However, at the sampling sites of GZ1-1, GZ4-2 and GZ4-3, low ^{240}Pu/^{239}Pu isotopic ratios of 0.147 ± 0.009, 0.080 ± 0.007 and 0.132 ± 0.044, respectively, were observed. In our previous study, we determined the vertical distributions of Pu isotopes in soil core samples from the

Table 2 Results of $^{239+240}$Pu activities and ^{240}Pu/^{239}Pu isotopic ratios in the surface soil samples

Sample	$^{239+240}$Pu activity (mBq g^{-1})[a]	^{240}Pu/^{239}Pu isotopic ratio[a]
JQ1	0.036 ± 0.004	0.186 ± 0.025
JQ2	0.049 ± 0.010	0.193 ± 0.053
JQ3	0.025 ± 0.009	ND
JYG	0.44 ± 0.02	0.186 ± 0.013
QQ1	0.38 ± 0.02	0.182 ± 0.012
QQ2	0.42 ± 0.01	0.189 ± 0.007
GZ1-1	0.74 ± 0.03	0.147 ± 0.009
GZ1-2	0.67 ± 0.11	0.167 ± 0.039
GZ3-1	0.054 ± 0.008	0.166 ± 0.032
GZ3-2	0.037 ± 0.014	ND
GZ3-3	0.19 ± 0.03	0.169 ± 0.033
GZ4-1	0.040 ± 0.008	0.182 ± 0.047
GZ4-2	0.19 ± 0.01	0.080 ± 0.007
GZ4-3	0.078 ± 0.017	0.132 ± 0.044
GZ4-4	0.68 ± 0.06	0.185 ± 0.024
DH1-S1	0.23 ± 0.02	0.165 ± 0.012
DH2-S1	0.87 ± 0.02	0.167 ± 0.006
DH2-S2	0.84 ± 0.10	0.179 ± 0.030
DH2-S3	0.89 ± 0.16	0.174 ± 0.042

ND Not detected

[a] uncertainties represent 1σ error

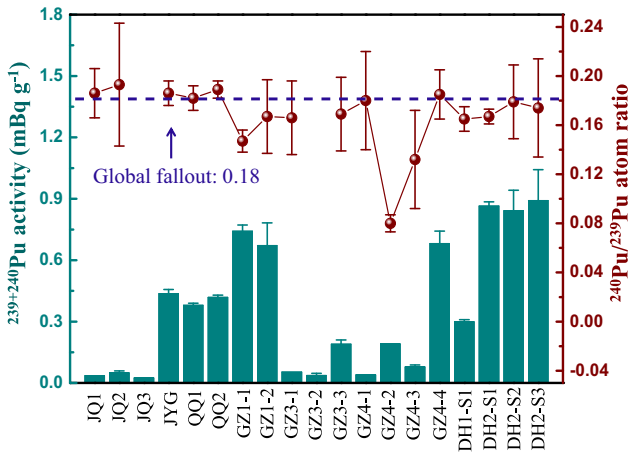

Fig. 3 Distribution of $^{239+240}$Pu activities and ^{240}Pu/^{239}Pu isotopic ratios in the surface soils from the Jiuquan region of Gansu province in northwestern China

Jiuquan region. The results showed that the ^{240}Pu/^{239}Pu isotopic ratios for the Pu isotopes from the CNTs recorded in the soil cores at the YM1 and YM2 sampling sites could be as low as 0.085 ± 0.002 and 0.059 ± 0.005, respectively (Fig. 1) [8]. The low ^{240}Pu/^{239}Pu isotopic ratios in the surface soil samples were comparable with the previous results. In Lake Bosten, which is located in the upwind area of the CNTs in Xinjiang province, a ^{240}Pu/^{239}Pu isotopic ratio of 0.080 ± 0.016 was also found in a sediment core at the depth of 5–6 cm and the low yield detonations of the CNTs conducted from 1967 to 1973 was suggested to be the source of this low ^{240}Pu/^{239}Pu isotopic ratio [30]. Zheng et al. [19] determined the vertical distribution of Pu isotopes in a soil core collected from Lanzhou city (Fig. 1) and the ^{240}Pu/^{239}Pu isotopic ratios were consistent with that of global fallout. Lanzhou city and the Jiuquan region are both in Gansu province with similar climate and geography conditions. The low ^{240}Pu/^{239}Pu isotopic ratios found in the surface soils further suggested that the Jiuquan region received regional fallout Pu from the CNTs and that the depostion from the CNTs was heterogeneous.

Regional fallout Pu from the CNTs

Investigating Pu contamination in the downwind areas from terrestrial nuclear test sites is important for radioactive source identification and radiological assessment. Compared with the Nevada nuclear test site and the Semipalatinsk nuclear test site, information about the distribution and characterization of Pu isotopes in the downwind areas especially the nearby downwind areas of the CNTs is limited up to date. The specific ^{240}Pu/^{239}Pu isotopic ratio, which could be used as fingerprint for Pu source identification, from the CNTs remains unknown. It has been reported that the ^{240}Pu/^{239}Pu isotopic ratio in the atmospheric samples collected after the 18st CNT was as high as 0.224 [31]. However, based on our results, this high ^{240}Pu/^{239}Pu isotopic ratio was not observed in the surface soils in the Jiuquan region. The 18st CNT was characterized by a high yield (4 Mt) and almost all of the produced radionuclides contributed to the stratospheric fallout [4]. Recently, Wu et al. [32] and Liao et al. [30] studied the distributions of Pu isotopes in the sediments from Lake Sugan in Gansu province and Lake Bosten in Xinjiang province in northwestern China. In the sediments of these two lakes, low ^{240}Pu/^{239}Pu isotopic ratios (near or below 0.1) were observed, revealing regional fallout from the CNTs. Small yield nuclear tests were conducted at the Nevada and Semipalatinsk test sites, resulting in low ^{240}Pu/^{239}Pu isotopic ratios (0.054–0.063 and 0.025–0.072 for the Nevada test site and Semipalatinsk test site, respectively) from these two nuclear test sites [5, 12]. Unlike these two nuclear test sites, the 22 atmospheric nuclear tests conducted at the CNTs were characterized by yields varying from 0.02 to 4 Mt. It is reasonable to assume that the related ^{240}Pu/^{239}Pu isotopic ratios reflected such yield variability and that, therefore, the Pu originating from the CNTs would present multiple fingerprints.

As part of our project focusing on the investigation of Pu isotopes in the soils from the downwind areas of the CNTs,

a previous study found that the $^{240}Pu/^{239}Pu$ isotopic ratios in the soils in the Jiuquan region were 0.059–0.186 with an average of 0.158 ± 0.011 [8]. Based on the calculation of the excess of the Pu inventory compared with that of global fallout, the contribution of Pu isotopes from the CNTs to this region was estimated to be more than 40 % [8]. The $^{240}Pu/^{239}Pu$ isotopic ratios (0.080–0.193) observed in the surface soils in the Jiuquan region further supported the finding that this region received fallout from the CNTs and that the mixing of such regional fallout with the global fallout resulted in $^{240}Pu/^{239}Pu$ isotopic ratios in some sampling sites slightly lower than that of the integrated global fallout.

Conclusions

In this study, Pu isotopes in the surface soil samples collected from the Jiuquan region in northwestern China have been determined. The $^{239+240}Pu$ activities varied with different sampling sites, suggesting the heterogeneous deposition of Pu in the Jiuquan region. The low $^{240}Pu/^{239}Pu$ isotopic ratios further supported the finding that this region received regional fallout Pu from the CNTs. The mixing of regional fallout Pu and global fallout Pu led to $^{240}Pu/^{239}Pu$ isotopic ratios in some sampling sites, slightly lower than the global fallout value.

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