Level, distribution and sources of plutonium in the coastal areas of China

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1 **Level, distribution and sources of plutonium in the coastal**
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Abstract: Plutonium is an important radioactive element in view of its biological toxicity and environmental impact. The two most important isotopes of plutonium, ^{239}Pu and ^{240}Pu , in the surface soil from the coastal area of China were determined using radiochemical separation combined with inductively coupled plasma mass spectrometry (ICP-MS) measurement. The average of the measured $^{240}\text{Pu}/^{239}\text{Pu}$ atomic ratios in all these samples is 0.186 ± 0.021 , which is in good agreement with the ratio of global fallout of the nuclear weapons tests. This indicates that the major source of plutonium in this region is global fallout, and there is no measurable release of plutonium from any nuclear power plant along the coast of China. The $^{239,240}\text{Pu}$ concentrations in all 71 soil samples range from 0.002 mBq/g to 0.670 mBq/g, which fall in the range of the reported values in the background area not directly influenced by nuclear activities. The anthropogenic disturbance of sampling sites, vegetation coverage of land, precipitation rate and organic substance content and erosion of soil have significant influence on the level of plutonium in the surface soil.

Keyword: Plutonium isotopes; source term of plutonium; spatial distribution; environmental impact; coastal areas of China

1. Introduction

Plutonium has been released to the environment by human nuclear activities, such as nuclear weapons tests, nuclear accidents, satellite accidents and nuclear fuel reprocessing plants (Qiao et al., 2009). Among them, the nuclear weapons tests released $6.5\times10^{15}\text{Bq}$ ^{239}Pu and $4.4\times10^{15}\text{Bq}$ ^{240}Pu , making it the dominant source of ^{239}Pu and ^{240}Pu in the environment (UNSCEAR, 1993). ^{239}Pu and ^{240}Pu are produced through sequential neutron activation reactions of uranium, this makes the $^{240}\text{Pu}/^{239}\text{Pu}$ atomic

ratios different in the samples with the different source of plutonium. The reported $^{240}\text{Pu}/^{239}\text{Pu}$ atomic ratios are 0.178 ± 0.023 for releases from global fallout of the nuclear weapons tests (Kelley et al., 1999), 0.01-0.07 for fuel of nuclear weapons (measured in the debris of plutonium fuel of nuclear weapons) (Eriksson et al. 2008), 0.23-0.67 for nuclear power plants (Warneke, 2002a; 2002b), 0.386-0.412 for releases from the Chernobyl accident (Boulyga et al., 1997; Wendt et al., 1999) and 0.323-0.330 for releases from the Fukushima accident (Zheng et al., 2013). This feature makes the $^{240}\text{Pu}/^{239}\text{Pu}$ ratio a unique fingerprint to identify the source of the radioactive substance in the environment and for nuclear forencics.

^{239}Pu ($T_{1/2}=24$ kyr) and ^{240}Pu ($T_{1/2}=6.5$ kyr) are the two most important isotopes of plutonium in the environment. Both of them are alpha emitters with high radiological toxicities, and therefore one of most important hazardous pollutant in the environment.

Based on the unique ratios of $^{240}\text{Pu}/^{239}\text{Pu}$ for the specific sources, plutonium isotopes in the environment have been used to investigate the source of radioactive substance and evaluate their environmental impact. The overall deposition of plutonium on the land from nuclear weapons tests and local releases of plutonium from nuclear activities makes plutonium isotopes a good tracer for investigation of soil erosion, redistribution of sediment, and water circulation in the ocean (Xu et al. 2013; Wang et al. 2017). An investigation on plutonium isotope in soil and sediment samples from the Liaodong Bay in northeast China and north Yellow Sea have been carried out to investigation of the soil erosion and input and transport of suspended particles/sediment in the seas (Xu et al. 2013, 2015, 2018). A level and distribution of ^{240}Pu and ^{239}Pu in soil in a large area in China have been investigated to identify the sources of the anthropogenic radioactive substances and their impact to the environemnt in China (Liaoning, Hebei, Shandong, Fujian, Guangxi, Guangdong, Zhejiang) (Xing, 2015;

Dong, 2010; Ni et al. 2018).

Rapid increased numbers of nuclear power plants were built in China in the past 20 years. To January 2019, 45 nuclear power reactors with the total capacity of 42.8 GW have been instilled and operated in China, and more than 11 nuclear power reactors are under construction (IAEA, 2019). All these nuclear power plants are located in the coastal area of China (Fig. 1). There is a big concern on the radiation impact of these nuclear power plants to the local environment. Although some investigation on the anthropogenic radionuclides including plutonium isotopes in the environment including the China have been reported, however, no systematic investigation of the anthropogenic radioactivity in the coastal area of China was reported.

This work aims to investigate the level, distribution and source of plutonium in the coastal areas of China, in order to assess the environmental impact of the nuclear power plants in this area, meanwhile to explore the influence of human activities and environmental and climate factors on the level and distribution of plutonium in the environment in the coastal area of China. This is going to be implemented by analyzing surface soil samples collected from this area for ^{239}Pu and ^{240}Pu using radiochemical separation and ICP-MS measurement.

2. Materials and Methods

2.1 Sampling

In total 71 surface soil samples were collected in depth of 0-5 cm (in most of sites) from the coastal area in China in July 2014, July 2015, April 2016 and April 2017 (Fig 2 and Table 1). The sampling sites were normally selected in a flat area without visible disturbance. However, the land in some areas in Beijing, Tianjin, Hebei, Shandong,

Jiangsu and Anhui province are highly cultivated by farming, it is difficult to find undisturbed sites. In this case, soil samples of 0-30 cm depth were collected using a stainless-steel soil core sampler of 7 cm in diameter and 30 cm in height. The surface soil samples of 5 cm depth were collected using a ring cutter of 7.0 cm in diameter and height of 5.0 cm. The samples from 5 points in a triangle of 1 m between each point were collected at each site and mixed as one sample. The collected soil samples were sealed in plastic bags and transported to the laboratory for analysis.

The soil sample was first weighted and air dried after removal of big stones (>5 mm) and vegetation roots. The samples were then dried in an oven at 150°C until constant weight. The dried samples were weighed, ground and sieved through an 80-mesh sieve.

2.2 Analytical method for determination of ^{239}Pu and ^{240}Pu in soil samples

The soil samples were analyzed for ^{239}Pu and ^{240}Pu using radiochemical separation combined with ICP-MS/MS measurement. A brief description of the analytical method is presented here, and the detailed analytical method has been reported elsewhere (Xing et al., 2018). The dried and ground soil was ashed at 450°C overnight, the loss of the sample mass (loss of ignition) was recorded as approximate mass of the organic substance in the sample. After spiked 5.0 pg of ^{242}Pu to the ashed sample as a chemical yield tracer, the sample was leached with 50 mL of *aqua regia* at 150°C for 0.5 hours and then 200°C for 2 hours. The leachate containing plutonium was separated from the residue by filtration through a glass fiber filter. Ammonium was added to the leachate to pH 8-9 to co-precipitate plutonium with $\text{Fe}(\text{OH})_3$. The precipitate was washed using 2 mol/L NaOH to remove amphoteric elements (such as Al, V, etc.). The precipitate was

then dissolved using HCl, and plutonium in the solution was reduced to Pu^{3+} by addition of $\text{K}_2\text{S}_2\text{O}_5$, and meanwhile iron was reduced to Fe^{2+} . Ammonium was added to pH 8-9 to co-precipitate $\text{Pu}(\text{OH})_3$ with $\text{Fe}(\text{OH})_2$, which was separated by centrifuge. The precipitate was dissolved with the concentrated HNO_3 , meanwhile Pu^{3+} was oxidized to Pu^{4+} with NO_2^- presented in the concentrated HNO_3 , and the solution was prepared in 1.0 mol/L HNO_3 . The prepared sample solution was loaded to a 2 mL of TEVA column which was conditioned using 1.0 mol/L HNO_3 . The column was rinsed with 1.0 mol/L HNO_3 and 6.0 mol/L HCl to remove the matrix and interfering elements. Plutonium adsorbed on the column was finally eluted with 0.1mol/L $\text{NH}_2\text{OH}\cdot\text{HCl}$ in 2mol/L HCl. The eluate of plutonium was evaporated to dryness, concentrated HNO_3 was added and evaporated to dryness again to decompose and remove $\text{NH}_2\text{OH}\cdot\text{HCl}$. The residue was dissolved with 0.5mol/L HNO_3 and transferred to a counting tube in a final volume of 3.5 mL. ^{239}Pu , ^{240}Pu and ^{242}Pu in the prepared sample solutions were measured using ICP-MS/MS (Agilent 8800 Agilent Technologies, Tokyo, Japan) using $\text{NH}_3\text{-He}$ as a reaction gas. The detection limits of this method for ^{239}Pu and ^{240}Pu were estimated to be 0.55 fg/mL for ^{239}Pu and 0.09 fg/mL for ^{240}Pu using the procedure blank in 3.5 mL HNO_3 solution. The blank was subtracted from the measured results of samples for calculation of the concentration of plutonium isotopes. A schematic diagram of the analytical procedure is shown in Fig. 3. It should be mentioned that the acid leaching method is suitable only for determination of global fallout plutonium in environmental samples such as soil and sediment. For plutonium present as refractory oxides, such as fallout in the close-in location of the nuclear weapons tests sites and nuclear accidents, a full dissolution method such as borate fusion and acid dissolution with HF is needed.

The above method was evaluated by analysis of soil (IAEA-327) and sediment

(IAEA-327) reference material. The measured concentrations of ^{239}Pu (0.349 ± 0.014 mBq/g) and ^{240}Pu (0.247 ± 0.011 mBq/g) and $^{240}\text{Pu}/^{239}\text{Pu}$ atomic ratio (0.192 ± 0.012) for IAEA-327 are in good agreement with the reference values for ^{239}Pu (0.35 ± 0.02 mBq/g) and ^{240}Pu (0.25 ± 0.02 mBq/g) and $^{240}\text{Pu}/^{239}\text{Pu}$ atomic ratio (0.19 ± 0.02) (Baeza et al., 2006). At the same time, the measured concentrations ^{239}Pu (1.887 ± 0.063 mBq/g) and ^{240}Pu (1.210 ± 0.044 mBq/g) and $^{240}\text{Pu}/^{239}\text{Pu}$ atomic ratio (0.175 ± 0.009) in IAEA-385 sediment are also agree well with the reference values for ^{239}Pu (1.79 ± 0.28 mBq/g) and ^{240}Pu (1.15 ± 0.14 mBq/g) and $^{240}\text{Pu}/^{239}\text{Pu}$ atomic ratio (0.174 ± 0.032) (Xing et al., 2018). Besides, the recoveries of plutonium measured by the spiked ^{242}Pu tracer range from 75% to 95%. The above results indicated that the analysis method for determination of trace concentrations ^{239}Pu and ^{240}Pu in environmental samples is reliable and accurate.

3 Results and discussion

3.1 Level and distribution of plutonium isotopes in the coastal areas of China

The concentrations ^{239}Pu and ^{240}Pu in 71 surface soil samples are shown in Table 1. Many data on the activity concentration of plutonium isotopes were reported for $^{239,240}\text{Pu}$ (sum of ^{239}Pu and ^{240}Pu), because the conventional method for determination of ^{239}Pu and ^{240}Pu is alpha spectrometry, which could not discriminate ^{239}Pu from ^{240}Pu due to their very close energy of alpha emissions. To easily compare with the reported values, the concentrations of $^{239,240}\text{Pu}$ in the analytical samples were calculated as $^{239}\text{Pu} + ^{240}\text{Pu}$. The results (Fig. 4) show that concentrations of $^{239,240}\text{Pu}$ are (0.002-0.670) mBq/g, with two orders of magnitude variation. The concentration of $^{239,240}\text{Pu}$ in soil in the background place without direct influence by nuclear activities have been reported to be (0.057-0.313) mBq/g in northeast China (Xing, 2015; Xu et al., 2015), (0.046-0.678) mBq/g in north China (Dong, 2010; Xing, 2015), (0.042-0.641) mBq/g in east

China, (0.033-0.173) mBq/g in south China (Xing, 2015), and (0.011-0.391) mBq/g in the similar latitude from other areas in China without human nuclear activity (Dong, 2010; Wang et al., 2013; Bu et al., 2014; Xing, 2015). The measured $^{239,240}\text{Pu}$ concentrations in this work fall into the reported range (0.042-0.678) mBq/g in China (Bu et al., 2014; Dong, 2010; Wang et al., 2013; Xing, 2015).

According to latitude of the sampling sites, average values of $^{239,240}\text{Pu}$ concentration in surface soil samples from each five-degree latitude band in a range of 20-45 °N are calculated to be 0.059 mBq/g in 20-25 °N, 0.117 mBq/g in 25-30 °N, 0.067 mBq/g in 30-35 °N, 0.108 mBq/g in 35-40 °N and 0.160 mBq/g in 40-45 °N band. Low $^{239, 240}\text{Pu}$ concentrations in the surface soil are observed in the latitude bands of 20-25 and 30-35 °N, and the highest area is measured in the band of 40-45 °N. This trend does not well agree with the reported distribution of the most anthropogenic radionuclides from the global fallout of the nuclear weapons tests (Hardy et al. 1973, UNSCEAR, 2000, Aoyama et al. 2006), in which a gradually increased concentrations of fallout radionuclides in the environment with the latitude in 20-45 °N were reported. The sites of the most atmospheric nuclear weapons tests were located in the mid-high latitude in the North Hemisphere (40-50°N). The radioactive substance was released to the troposphere and injected to the stratosphere. The debris released to the troposphere is mainly deposited locally, especially for the relatively large particles forming close-in deposition, gaseous radionuclides and small particles transported to the upper troposphere were dispersed to a relative larger distance. The radioactive substances injected to the stratosphere and upper troposphere are mainly from large explosion equivalent explosion, especially those thermonuclear devices, these substances were mainly dispersed in the similar latitude due to the rotation of the earth, although some fraction in the stratosphere was also dispersed cross the latitude, even cross the

hemisphere. All these caused a high deposition of the fallout of nuclear weapons test in the middle and high latitude in the North Hemisphere. Such distribution trend of global fallout has been observed all over the world (UNSCEAR, 2000).

The lowest $^{239,240}\text{Pu}$ concentrations in surface soil (0.003-0.116 mBq/g with an average of 0.059 mBq/g) was observed in the area of the lowest latitude of 20-25 °N, and highest $^{239,240}\text{Pu}$ concentrations (0.031-0.670 mBq/g with an average of 0.160 mBq/g) in the area of the highest latitude of 40-45 °N in the entire investigation area of 20-45 °N. This variation follows the general distribution of the global fallout of the nuclear weapons tests all over the world (UNSCEAR, 2000). However, a relatively low $^{239,240}\text{Pu}$ concentrations in surface soil (0.002-0.122 mBq/g with an average of 0.067 mBq/g) was observed in the area of 30-35°N compared to those observed in the lower latitude area in 25-30°N (0.004-0.223 mBq/g with an average of 0.117 mBq/g) and the higher latitude area in 35-40°N (0.009-0.151 mBq/g with an average of 0.108 mBq/g). This might be attributed disturbance of human activity in these sites. The soil samples in the latitude band of 30-35°N were mainly collected from agriculture land because of high cultivation in this area and difficulty to find undisturbed site. In these sites, the deposited plutonium was extensively mixed in the top 30 cm layer during the cultivation activity, and therefore the upper 30 cm soil was collected for analysis (Table 1). A relatively homogenous distribution of anthropogenic radionuclides (^{137}Cs , ^{239}Pu , ^{240}Pu) has been reported in the agriculture land because of the periodic mixture of the soil (Xu et al. 2015), this caused a low concentration of $^{239, 240}\text{Pu}$ in the surface soil collected from the cultivated land compared to those from the undisturbed sites (Xu et al., 2015).

Relatively high $^{239,240}\text{Pu}$ concentrations were observed in the surface soil collected in the area in 25-30°N (0.004-0.223 mBq/g with an average of 0.117 mBq/g) compared to those collected in the area of higher latitude of 35-40°N (0.009-0.151 mBq/g with an

average of 0.108 mBq/g). This might be attributed to the vegetation coverage and organic substance content in the soil. The sampling sites in the area of 25-30°N are mainly located in forest with high vegetation coverage, which prevents from the erosion of the soil, therefore well reserved plutonium in the soil (Li et al., 2014). Relative higher organic content (ignition loss) were measured in the samples in the area of 25-30°N (4-8%) compared to the sites of other areas. The fractionation analysis has shown that large fraction of plutonium is associated with organic substances in the soil (Qiao et al. 2012). mainly in a stable complex with humic and fulvic acids in soil (Lee et al. 1997). The high content of organic substance can help the fixation of plutonium after it deposited and entered the soil, and reduce the migration of plutonium the deep soil.

The highest $^{239,240}\text{Pu}$ concentration of 0.670 mBq/g was observed in the sample S14152, which was collected at Zhangjiakou in Hebei province (41.2 °N, 116.2 °E) with a precipitation rate of 385 mm/yr. It has been reported that both gaseous and particle associated radionuclides can be effectively trapped and washed out from the atmosphere to be deposited to the earth, therefore wet deposition is the major deposition of radionuclides from the atmosphere (UNSCEAR, 2000). Combined with a relatively high latitude of this site (41.2 °N), corresponding to the high level of plutonium released during the atmospheric nuclear weapons test in 1945-1980 mainly in the high latitude of North Hemisphere, a high fallout level of anthropogenic radionuclides from the nuclear weapons tests is expected, causing a high $^{239,240}\text{Pu}$ level in this site. A similar high $^{239,240}\text{Pu}$ concentration in surface soil (0.687mBq/g) was also reported in Beijing, in the same latitude in Beijing (Dong, 2010). In addition, this sampling site is located in a forest, and a high organic substance content of 11.9% was measured in this soil, therefore a less loss of plutonium by soil erosion and less migration of plutonium in the surface soil are expected in this site, which helps to reserve the deposited plutonium in

the surface soil in this site.

The lowest $^{239,240}\text{Pu}$ concentrations were observed in soil sample S15097 (0.002 mBq/g) collected at Taizhou in Jiangsu province (32.3 °N, 120.0 °E) with a relative high precipitation rate of 1022 mm/yr (Xu, 2017) and S16016 (0.002 mBq/g) collected at Shangrao in Jiangxi province (28.4 °N, 117.7 °E) with a relatively high precipitation rate of >1500 mm/yr. These two samples are sandy soil with very low organic substance content (1.1 % and 2.7%) and the two sites were also less covered by vegetation, all these features cause a high loss of surface soil by erosion, and less fixation of plutonium resulting in a high migration of plutonium from the surface soil. Combined with the less deposition in the low latitude area, a low $^{239,240}\text{Pu}$ concentration occurred in these soil samples.

3.2 Sources of plutonium in the environment in the coastal area of China

$^{240}\text{Pu}/^{239}\text{Pu}$ atomic ratios in the surface soil samples collected in the coastal area of China were measured to be 0.154-0.227 with an average of 0.186 (Fig. 5), which is in a good well agreement with the reported ratio of global fallout of nuclear weapons tests (0.178 ± 0.023) (Kelley et al., 1999), but significantly different with the values in the fuel of nuclear weapons and the close-in deposition of nuclear weapons tests (0.01-0.07) (Eriksson et al. 2008), releases from the Chernobyl accident (0.386-0.412) (Boulyga et al., 1997; Wendt et al., 1999), Fukushima accident (0.323-0.330) (Zheng et al., 2013) and some NPP (0.23-0.67) (Warneke, 2002a). These results indicate that the plutonium in the surface soil in the study area is dominantly originated from the global fallout of the nuclear weapons tests in 1945-1980.

Nuclear accident in Chernobyl happened in April 1986 had released a large amount of radioactive substances into the environment, besides the deposition in the local

region, the radioactive plume was also dispersed to large area and deposition mainly in the Europe (Levi, 1991; Muramatsu et al., 2000). Meanwhile, low level of Chernobyl derived ^{131}I and ^{137}Cs were also measured in China, mainly in North China. Although some amount of plutonium was also released from the Chernobyl nuclear accident and observed in the heavy contaminated area in Ukraine, Belorussia and Russia (UNSCEAR, 2000; Thakur et al. 2017). No Chernobyl derived plutonium was reported in China and the very low level of Chernobyl derived plutonium was observed in the air sample in Japan in a short period after the Chernobyl accident (Hirose et al. 1994), because plutonium released from the Chernobyl was mainly associated with particles, and deposited in the close-in place. Fukushima nuclear accident happened in 2011 had also released a mass of radioactive substances into the environment, but plutonium derived from Fukushima nuclear accident was detected only in some samples around Fukushima Daiichi NPP (<20 km) (Zheng et al., 2013), no Fukushima derived plutonium was observed in other region including China. In addition, only lower level of Fukushima derived ^{131}I and ^{137}Cs was detected in China during the accident due to the dominant westwards wind. Therefore, the contribution of two nuclear accidents to plutonium in the soil samples in the study area can be neglected.

The sampling sites are more than 2000 km far to the nearest nuclear weapons tests site at Lop Nor. Although some radioactive substances released from the weapons tests at this site in 1964-1980 might be transported and deposited in the sampling site due to the dominant westerlies in North China, the contribution of the close-in deposition of plutonium at Lop Nor to plutonium in the sampling region might be limited because of the particles associated feature and long distance. The $^{240}\text{Pu}/^{239}\text{Pu}$ atomic ratios, reported in the soil from other area in China, such as Shangqiu (0.186-0.218) (Wang et al., 2013), Puyang (0.179-0.189) (Wang et al., 2013), Jingmen (0.170-0.211) (Dong,

2010), Xiangyang (0.186-0.220) (Dong, 2010), Changzhi (0.143-0.215) (Wang et al., 2013), Lanzhou (0.168-0.192) (Zheng et al., 2009) and Chengdu (0.184-0.223) (Dong, 2010) confirmed no detectable effect of plutonium in soil in the background area in China. The $^{240}\text{Pu}/^{239}\text{Pu}$ atomic ratios in the sediment from Lake Hongfeng (0.162-0.213) (Zheng et al., 2008b), Lake Poyang (0.185-0.192) (Liao et al., 2008), Lake Shuangta (0.160-0.192) (Wu et al., 2010), Lake Chenghai (0.164-0.223) (Zheng et al., 2008a), Dianchi (0.164-0.187) (Shi, 2015) and Lake Fuxian (0.173-0.201) (Shi, 2015) in China also showed no detectable influence on the plutonium deposition in these areas. These data suggest that the close-in deposition of Chinese nuclear weapons tests have a limited influence on the plutonium in the soil in the downwind area far away from nuclear activity areas.

The analytical results of $^{240}\text{Pu}/^{239}\text{Pu}$ ratio in the surface soil collected in the coastal areas of China and the above discussion indicate that except the global fallout of atmospheric nuclear weapons tests before 1980, no other source of plutonium in this region is detectable. This also implies that no detectable plutonium was released from the nuclear power reactors along the coastal areas of China, confirming the safe operation of these nuclear power plants.

4. Conclusion and perspectives

Based on the results and discussion above, it can be concluded: (1) The concentrations of $^{239,240}\text{Pu}$ in the surface soil in the coastal area of China ranging from 0.002 mBq/g to 0.670 mBq/g is comparable to the values reported in other locations in China and other similar latitude place of background areas. The distribution of $^{239,240}\text{Pu}$ concentration in the surface soil depends on the sampling site related to the deposition level of anthropogenic radionuclides due to the most of nuclear weapons tests sites located in

the middle and high latitude area in the North Hemisphere. The vegetation coverage and organic substance content in the soil help the reservation of deposited plutonium in the surface soil. (2) Plutonium in the environment in the coastal area of China is dominantly originated from the global fallout of atmospheric nuclear weapons tests with the typical $^{240}\text{Pu}/^{239}\text{Pu}$ ratio of 0.18, contribution of the other sources to the plutonium in this region is negligible. (3) There is no detectable effect of the NPPs along the coast of China to the environment, in view of the plutonium. The analytical results of this work provide valuable baseline values of plutonium isotopes in the environment in the coastal area of China, which will be very useful for the evaluation of the impact of NPPs along the coast of China because of the rapid increased installation and operation of nuclear power plants in this region in the past years as well as in the coming years.

Declaration of interest

None.

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Table 1 Analytical results of ^{240}Pu and ^{239}Pu concentration in the surface soil samples collected from coastal areas in China

Caption of figures

Fig. 1 Location of nuclear power plants and nuclear power units in operation and under construction in China until January 2019 (based on the data in IAEA, 2019)

Fig. 2 Sampling sites of surface soil samples in the coastal areas of China

Fig. 3 Schematic diagram of analytical procedure for determination of Pu isotopes in soil samples

Fig. 4 Distribution of $^{239, 240}\text{Pu}$ concentrations in surface soil in the coastal areas of China

Fig. 5 Distribution of $^{240}\text{Pu}/^{239}\text{Pu}$ atomic ratio in surface soil in the coastal areas of China