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Plutonium isotopes in the Qinghai-Tibet Plateau: Sources, distribution, and their environmental behaviors

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\textbf{ABSTRACT}

Due to the high radiotoxicity in high concentrations, plutonium isotopes have drawn high attentions in the consideration of radiation risk, their sources, level, environmental behaviors, including deposition, retention and migration behaviors. However, such research in the Qinghai-Tibet Plateau is still missing, where is deemed as an environmental sensitive area. \textsuperscript{239,240}Pu in surface soil collected from the Qinghai-Tibet Plateau were determined for the first time in this work. The concentrations of \textsuperscript{239,240}Pu are in the range of 0.0176–1.95 Bq/kg, falling into the reported ranges in the background areas from the similar latitude belt. The \(240\text{Pu}/239\text{Pu}\) atomic ratio range was measured to be 0.146–0.225, which is similar with the global fallout values. Both indicate that the global fallout is the major source of plutonium in this region, and the low plutonium level will not cause any radiation risk so far. Based on the statistical analysis of the possible parameters (organic content, moisture content, average annual precipitation, altitudes, topography and human activity), the large variations of \textsuperscript{239,240}Pu concentrations were mainly attributed to the retention process related factors including soil organic content and human activity disturbances. While, the deposition related factors including the average annual precipitation, altitudes, topography made insignificant influence on the spatial distribution of \textsuperscript{239,240}Pu concentrations due to the low \textsuperscript{239,240}Pu concentrations in atmosphere, less wet deposition amount and insignificant re-suspended amount. The highest \textsuperscript{239,240}Pu concentrations of 0.805–1.95 Bq/kg were mainly due to the good retention condition in the sampling sites with higher soil organic content and less human activity disturbances.

\section{1. Introduction}

Over 95\% of the anthropogenic plutonium (Pu) in the environment is originated from the global fallout of atmospheric nuclear weapons tests (NWTs) during 1950–1960s, which injected large amounts of radioactive isotopes including plutonium into the stratosphere. After mixing, these radionuclides deposited all over the world, especially in the mid-high latitude region of the Northern Hemisphere. Besides that, low-yield atmospheric NWTs, operating of nuclear fuel reprocessing plants (NFRPs) and other nuclear facilities (e.g. nuclear power plants and nuclear research facilities), and nuclear accidents also released some amount of plutonium to the environment, which is mainly deposited in the nearby areas within several hundred kilometers (Alewell et al., 2017). Due to the high radiotoxicity in high concentration of \(\alpha\) emitting \textsuperscript{239}Pu (\(T_{1/2} = 24,100\) yr) and \textsuperscript{240}Pu (\(T_{1/2} = 6561\) yr) (Zhao et al., 2020), the level and sources of \textsuperscript{239,240}Pu in the environment is of great significance in the view of radiation risk. Meanwhile, the environmental behaviors, including deposition, retention and migration of plutonium are also important in the investigation of their potential hydrosphere contamination, uptake by flora and fauna and transfer to food chain (Kirchner et al., 2009).

As one of the most important sinks of plutonium, a lot of effort has
been paid to survey the $^{239,240}$Pu level in surface soil in China, including several early investigations in the surrounding areas of Chinese NWTs site- Lop Nor in the 1980s and 1990s (Pan et al., 1996; Ren et al., 1998; Zhang et al., 1997), and in Northwest (Bu et al., 2015; Zhao et al., 2020; Zhang et al., 2019), North (Dang et al., 2021; Xing, 2015; Cao et al., 2019), Northeast (Xu et al., 2013; Zhang et al., 2016; Zhang et al., 2021), Central (Dong et al., 2010; Wang et al., 2013), Southwest (Bu et al., 2014; Ni et al., 2018), and South China after 2010 (Ni et al., 2020; Zhang and Hou, 2019). However, the investigation of plutonium in the Qinghai-Tibet Plateau is still missing. Qinghai-Tibet Plateau acts as the largest fresh water reservoir in Asia (including over $2 \times 10^{11}$ m$^3$ of glacier resource, $3 \times 10^{10}$ m$^3$ of annual runoff, and 1800 lakes), has a unique environment (an average altitude of 4000 m, 8 main mountain ranges, annual precipitation rate ranged from <100 mm to >1000 mm, and diversified landscapes including mountainous area, Gobi Desert, pastureland, glacier, etc.), and deemed as an environmental sensitive area.

This work aims to investigate the level, sources and distribution of plutonium in surface soil from the Qinghai-Tibet Plateau, and explore the deposition and retention behaviors of plutonium in such a special environment, in order to better understand for the radiation safety status in this area.

2. Materials and methods

2.1. Studied area and sampling

The investigated area is in 27.5°-36.4° N, 79.9°-106.5° E (Fig. 1), which stretches across most areas of the eastern Tibetan Plateau and south margin of the plateau. Due to the hard accessibility to the north-west region, only a few samples were collected from there. The investigated area is mainly consisted of the pasture land, Gobi Desert, mountains (Hengduan Mountains, Bayan Har Mountains, Tanggula mountains, etc.) (Fig. 1), and the altitudes (470 m-4797 m, mean: 3478 m), annual average temperatures (~5.6 °C-16 °C), and annual average precipitation (66–1031 mm, mean: 531 mm) varies in a large range (http://www.cma.gov.cn).

Surface soil samples (0–5 cm) were collected at 65 sites with a ring cutter of 6.8 cm in inner diameter and height of 5.0 cm in Aug. 2019 (Fig. 1). Most samples were collected far away from artificial disturbed sites, including in pastureland, desert, mountains, etc. However, samples nearby the agriculture land and roads were also unavoidably collected, especially in Sichuan Basin and on the single route to get into the plateau. At each site, samples from 3 points in a triangle of 1 m distance were collected and mixed as one. The collected soil samples were sealed in plastic bags and transported to the laboratory for analysis. The sample was first air dried after removal of gravels and vegetation roots, and then dried in an oven at 60 °C until constant weight. The sample weight was recorded both before and after drying to calculate the soil moisture content. The dried samples were then ground and sieved through an 80-mesh sieve.

2.2. Analysis of soil samples for plutonium isotopes

The plutonium isotopes ($^{239}$Pu, $^{240}$Pu) in soil samples were determined using the procedure modified from Qiao et al. (2009). In short, an aliquot of 5 g soil was weighed into a beaker and ashed in a muffle furnace at 450 °C for 12 h to decompose organic matters. The loss of ignition (LOI) was calculated based on the weight loss before and after ashing. The $^{242}$Pu of 1 mBq was spiked as tracer to monitoring the chemical yield of plutonium in the chemical separation procedure. Aqua regia was used to digest samples and leach plutonium from soil particles. After filtration, ammonia was added to the leachate to co-precipitate plutonium with Fe(OH)$_3$ at pH = 8–9. The precipitate was dissolved...
with conc. HCl, and all the plutonium isotopes were then reduced into Pu$^{3+}$ by addition of K$_2$S$_2$O$_5$ and precipitated again by adjusting pH = 9–10 with ammonia. The precipitate was separated and dissolved with conc. HCl, and conc. HNO$_3$ was added to oxidized Pu$^{3+}$ to Pu$^{+}$. The sample was prepared in 1 M HNO$_3$ solution and loaded to a TEVA column (2 ml). After rinsing with 120 ml of 1 M HNO$_3$ and 60 ml of 6 M HCl to remove interfering elements (U, Th), plutonium on the column was finally eluted with 40 ml of 0.1 M NH$_3$OH-HCl-2 M HCl solution. The eluate was evaporated to dryness, a few drops of conc. HNO$_3$ was added and evaporated to dryness to decompose the hydroxylamine and eliminate the remained HCl. The residue was dissolved in 0.5 M HNO$_3$. Samples collected from the same small region were analyzed together to avoid cross contamination. For quality assurance, replicate analysis of two samples (S19213 and S19024) were implemented, the results of two analyses of the same sample agreed well. Each batch analysis consists 7 sample with one blank, to evaluate the possible contamination and blank subtraction.

$^{239,240}$Pu, $^{240}$Pu, $^{242}$Pu in the separation solution were measured using a triple quadrupole ICP-MS (Agilent 8800 ICP-MS/MS, Agilent Technologies, Tokyo, Japan) with NH$_3$He mixtures gases as the reaction gas. The details about the measurement method have been reported elsewhere (Xing et al., 2015). The detection limits of this method for $^{239}$Pu and $^{240}$Pu were estimated to be 0.55 fg/ml and 0.09 fg/ml, respectively, using 3 times standard division of the count rates of the procedure blanks and the measured sensitivity of instrument for plutonium isotopes. The measured signal intensities of $^{239}$Pu and $^{240}$Pu in procedure blanks were 0.5–0.6 cps and 0.04–0.07 cps, respectively, which were 1–3 orders magnitudes lower than that in samples, and subtracted from the samples in the same batch. $^{240}$Pu spiked to the sample before separation was used as an isotope dilution standard to calculate the concentrations of $^{239}$Pu and $^{240}$Pu in the samples, as well as to estimate the recoveries of plutonium in the chemical separation (most are better than 75%).

3. Results

The concentrations of $^{239,240}$Pu (sum of $^{239}$Pu and $^{240}$Pu) in 65 surface soil samples vary from 0.0176 to 1.95 Bq/kg with a mean of 0.345 ± 0.032 Bq/kg (Fig. 2). Seven highest values from 0.805 to 1.95 Bq/kg were observed in Nyingchi Prefecture (n = 2) and Changdu area (n = 1) in Tibet, Ganzi county (n = 3) in Sichuan province, and Guoluo Prefecture in Qinghai province (n = 1). Relatively low $^{239,240}$Pu concentrations (<0.118 Bq/kg, n = 19), i.e. less than 1/3 of the mean, were mainly measured in the interior of plateau (average altitude >4100 m), such as in Shigatse Prefecture, Nagqu Prefecture, Ngari Prefecture and Lhasa Area. Beyond that, low values also scattered in the east and middle of studied area, which were measured in farmland and next to the road (Table S1).

The measured $^{240}$Pu/$^{239}$Pu atomic ratios were in the range of 0.146–0.225 with an average of 0.185 (Fig. 2). About 32% of the samples were collected in 25–30’N, and the remaining 68% in 30–35’N. The measured ratio range is similar with the reported global fallout values from the same latitude belt (0.178 ± 0.019 in 0-30’N and 0.180 ± 0.014 in 30–71’N) (Kelley et al., 1999).

4. Discussion

4.1. Sources of $^{239,240}$Pu in the Qinghai-Tibet Plateau

The measured $^{239,240}$Pu concentrations (0.0176–1.95 Bq/kg) in the surface soil from the Qinghai-Tibet fall into the reported range in the background areas worldwide (0.37–3.70 Bq/kg) (ATSDR, 2010), especially in the similar latitude areas, such as 0.005–2.66 Bq/kg in the mainland of China (Guo et al., 2019), 0.02–1.85 Bq/kg in neighboring countries (Japan and Korea) (Lee and Lee, 1997; Hirose et al., 2004), and 0.71–4.69 Bq/kg from the Alps mountains (Alewell et al., 2014). The measured $^{240}$Pu/$^{239}$Pu atomic ratios of 0.146–0.225 with an average of 0.185 are similar to the reported ratios of global fallout of the atmospheric nuclear weapons tests (Fig. 3), i.e. 0.178 ± 0.019 in 0-30’N and 0.180 ± 0.014 in 30–71’N (Kelley et al., 1999), as well as to the reported values in the background areas of China where only receive global fallout originated $^{239,240}$Pu, such as in the coastal area (0.154–0.227), northeast (0.144–0.245), north (0.143–0.218), and south of China (0.165–0.202) (Zhang and Hou, 2019; Xu et al., 2013; Wang et al., 2013; Ni et al., 2020). Meanwhile, the measured $^{240}$Pu/$^{239}$Pu atomic ratios are clearly different from the values observed in the releases of the Chernobyl accident (0.403–0.412), Fukushima accident (0.323–0.330), nuclear power plants (0.23–0.67) and the fuel of nuclear weapons (0.01–0.07) (Boulyga et al., 1997; Zheng et al., 2012; Warneke et al., 2002). The lowest $^{240}$Pu/$^{239}$Pu ratio of 0.146 ± 0.015 and highest ratio of 0.225 ± 0.015 were measured in the samples with low $^{239}$Pu and $^{240}$Pu concentrations, besides the high measurement uncertainty of the low $^{240}$Pu signal which caused a relatively high analytical uncertainties for $^{240}$Pu/$^{239}$Pu atomic ratios, inhomogeneous deposition of plutonium from different nuclear weapons tests could not be excluded. However, no clearly geographical pattern of $^{240}$Pu/$^{239}$Pu atomic ratios in the study area was observed in this work, this might be partly attributed to insufficient number of samples analyzed and no data on depth profile of plutonium isotopes, a further comprehensive investigation is needed for clarification of this issue. Anyway, $^{239,240}$Pu in the

Fig. 2. Descending $^{239,240}$Pu concentrations in surface soil in Qinghai-Tibet Plateau and the corresponding $^{240}$Pu/$^{239}$Pu atomic ratios.
studied soil samples should originate mainly from the global fallout of atmospheric nuclear weapons tests in 1945–1980.

Plutonium was seldom released into the atmosphere from nuclear fuel reprocessing plants (NFRPs) due to its high boiling point and not volatile at relative high temperature (\(<1000\) °C) (Zhao et al., 2020). The two largest NFRPs at Sellafield (UK) and La Hague (France) are far away from the investigated area. Although the nearest sampling site is only \(~500\) km from the small Chinese NFRPs in Jiayuguan, Gansu Province, the Qilian Mountains with an average altitude of 4000–5000 m lie in the middle of the sampling area and this reprocessing plant, and no distinct \(^{240}\text{Pu}/^{239}\text{Pu}\) ratio (mean: 0.167 \(\pm\) 0.013) were measured in the area nearby this NFRPs (Xing, 2015). Several sampling sites are also very close to another small Chinese NFRPs in Guangyuan, Sichuan Province (\(~100\) km), and no distinct \(^{240}\text{Pu}/^{239}\text{Pu}\) ratio (0.173 \(\pm\) 0.011) was measured as well. The released \(^{239,240}\text{Pu}\) might be well limited in a relatively small range (\(<10\) km) due to the complex mountainous terrain near these two NFRPs. The nuclear accidents at Chernobyl and Fukushima have released a small amount of plutonium to the environment. While, the particle associated plutonium released from Chernobyl accident was mostly deposited in the local areas, such as in Ukraine, Belorussia and Russia (UNSCEAR, 2000), and no Chernobyl derived plutonium has been detected in China. About 4 orders of magnitude less amount of \(^{239,240}\text{Pu}\) were released from Fukushima compared to that of Chernobyl, and most released plutonium was only detected in soil samples collected in the 30 km exclusive zone of accident (Zheng et al., 2012).

The two nearest nuclear weapons testing sites at Lop Nor (900–1600 km) and Semipalatinsk (2200–3000 km) have also released a certain amount of plutonium to the atmosphere, including troposphere from those low-yield nuclear weapons tests and resulted in close-in deposition in the local area nearby the testing site. The big difference of the explosion yields of these weapons from 0.02 Mt to 4 Mt TNT might cause different \(^{240}\text{Pu}/^{239}\text{Pu}\). However, the radioactive clouds of the tests at Lop Nor were mainly dispersed southeast, east and northeast direction (Bu et al., 2015), not south direction to the studied area. Meanwhile, no clear trend of \(^{240}\text{Pu}/^{239}\text{Pu}\) ratios and \(^{239,240}\text{Pu}\) concentrations in the downwind areas of Lop Nor was observed (Xing, 2015). Large numbers of nuclear weapons tests conducted at the Semipalatinsk Nuclear test site where is located in the northwest of studied area, and causing a close-in deposition of plutonium with relatively low \(^{240}\text{Pu}/^{239}\text{Pu}\) ratios (0.025–0.072). The reported \(^{240}\text{Pu}/^{239}\text{Pu}\) ratios in soil samples from Northern Xinjiang, the closest area of China to the Semipalatinsk, have shown Semipalatinsk derived plutonium signal in the northwest corner of Northern Xinjiang, but not outside of this region (Zhao et al., 2020). Therefore, the close-in fallout of \(^{239,240}\text{Pu}\) released from these two sites should not be a significant source in the studied area.

There are no nuclear power plants and other nuclear facilities operating in this region until now, and most Chinese nuclear power plants are in the east coast area with no distinctly elevated \(^{239,240}\text{Pu}\) concentrations and abnormal \(^{240}\text{Pu}/^{239}\text{Pu}\) ratios measured in surface soil samples from there (Zhang and Hou, 2019). Therefore, \(^{239,240}\text{Pu}\) in the studied area was mainly originated from the global fallout (from stratosphere) of the atmospheric nuclear weapons tests.

### 4.2. Distribution of \(^{239,240}\text{Pu}\) in surface soil in the Qinghai-Tibet Plateau and the main influence factors

The \(^{239,240}\text{Pu}\) concentrations in the surface soil from the studied area varied by a factor of \(~100\) (0.0176–1.95 Bq/kg). The seven highest values from 0.805 to 1.95 Bq/kg were observed in Nyingchi Prefecture (n = 2) and Changdu area (n = 1) in Tibet, Ganzi county (n = 3) in Sichuan province, and Guoluo Prefecture in Qinghai province (n = 1). According to the comparable \(^{240}\text{Pu}/^{239}\text{Pu}\) ratios (0.181–0.193, mean: 0.189 \(\pm\) 0.014) in these seven samples with that of global fallout (0.178
\( \pm 0.019 \) in 0-30\(^{\circ}\)N and 0.180 \( \pm 0.014 \) in 30–71\(^{\circ}\)N, no other source of plutonium in these sites are expected. High deposition and retention of plutonium in these sites should be the main reason for their high plutonium concentrations. All these sites are in mountainous region and pasture land with relatively higher vegetation coverages (field records), higher annual precipitation rate (mean: 637 mm) and less human activities. The LOI values (mean: 22\%) and moisture content (mean: 31\%) in these soil samples are higher as well. The highest concentration of \(^{239,240}\)Pu of 1.95 \( \pm 0.08 \) Bq/kg was measured in a soil sample collected in the undisturbed pasture land. The inventory of this sample was calculated to be 88.1 Bq/m\(^2\) based on the dry weight, sampling volume and sampling depth (5 cm), which is comparable with the reported values (84.5–138 Bq/m\(^2\)) measured from the whole undisturbed soil profiles (0–30 cm) in the similar latitude belt (Zhang et al., 2019; Zhao et al., 2020). Meanwhile, this soil also showed a LOI value as high as 35\% and a relatively humid climate was recorded nearby (annual mean precipitation of 648 mm), therefore, the lush vegetation and a well retention of plutonium mainly in the surface 5 cm of soil could be indicated in this site.

While relatively low plutonium levels (<0.118 Bq/kg, \( n = 19 \)) were mainly observed in surface soils collected from the interior of Qinghai-Tibet Plateau with an average altitude >4200 m, in Shigatse Prefecture, Nagqu Prefecture, Ngari Prefecture and Lhasa area. Most of these sites are in Gobi Desert and mountainous areas with low vegetation coverages, and precipitation rates (mean: 321 mm), and these soils have low LOI values (mean: 5\%) and moisture content (10\%). Beyond that, low values also scattered in the east and middle of studied area. Because of the high agriculture activities and difficulties to find un-disturbed sites in these areas, these soil samples were unavoidably collected from artificial disturbed areas such as farm land and next to the road (Fig. 4).

The level of Pu in soil is determined by two stages/processes, deposition of Pu from atmosphere and retention of Pu in the soil after deposition. Since the vegetation coverage is highly influenced by the annual precipitation rate and soil moisture content, which is manifested as soil organic content in return. Therefore, only the annual precipitation rate, soil moisture content and organic content are considered as the possible factors, of which the annual precipitation rate could affect the deposition of plutonium, and soil moisture and organic content could affect the retention of plutonium in the soil. The topography and altitude have been reported to be important in the deposition of airborne pollutants (Kang et al., 2019; Zhao et al., 2020), which could also be possible factors influencing the plutonium concentrations in surface soil samples as well. All these factors together with the human activity (could affect the retention of plutonium as reported) could interact with each other and determine the final distribution of plutonium concentrations in the studied surface soil samples (Zhao et al., 2020).

A simple statistical analysis of these possible parameters (organic content, moisture content, average annual precipitation, altitudes, topography and human activity) is implemented to explore the main factors governing the deposition and retention of plutonium in the soil from the Qinghai-Tibet Plateau. Here the multiple linear regression model was applied, therefore, the linear correlation between concentrations and each factor were first analyzed to determine the linear relation rather than other kinds of relations.

The organic content was presented by the LOI value and measured to be 0.33\%–40\% with a mean value of 10\%. The moisture content was measured to be 0.06–92\% with a mean value of 17.5\%. The average annual precipitation of 66–1031 mm with a mean value of 531 mm was obtained at or near each site from the website of China Meteorological Administration (http://www.cma.gov.cn.). The altitudes measured at each sampling site vary in 470–4797 m with a mean value of 3478 m. For the rest two non-numerical parameters, human activity and topography, a simple parametric process was done based on the reported
physical significance on the concerned plutonium concentrations (Zhao et al., 2020; Zhang et al., 2021; Lee and Lee, 2001). A value of 1 was assigned to the sample collected at sites disturbed by human activities, and 2 for the samples from sites without human disturbance. A value of 2 for the samples collected from mountainous areas and 1 for that collected from other types of topography (Table S1).

The individual linear regression analysis shows significant correlations between the concentrations of $^{239,240}$Pu and LOI ($r = 0.71$), soil moisture content ($r = 0.52$), and human activities ($r = 0.37$) ($\alpha = 0.05$, $n = 63$, critical value = 0.24), while no significant linear correlations between the plutonium concentrations and annual precipitation rate ($r = 0.19$), altitude ($r = 0.08$), and topography ($r = 0.18$) were found when $\alpha = 0.05$. This might imply that LOI (organic matter) and moisture content in the soil and human activities of sampling sites could be the key parameters influencing the distribution of plutonium in the studied area, therefore, these three parameters were further analyzed with the multiple linear regression model. Table 1

As results, Multiple R is 0.77 ($\alpha = 0.05$, $n = 65$-3-1, critical value = 0.25), which is higher than any of the $r$-value for each single factor, indicating a better results of the multiple linear regression model. The $R^2$ is 0.59, and significance F is $6.78 \times 10^{-12}$ ($<0.05$), the whole regression equation passed the F-test. The t Stat of two factors, including LOI and human activities, is 5.48, 3.67 (absolute value > 2), and P-value is 8.52 $\times 10^{-7}$ and 5.05 $\times 10^{-5}$ ($<0.05$), respectively, which passed the t-test. Therefore, the factors mainly related with the retention process (the LOI and human activities), should be the key factors affecting the final distribution of $^{239,240}$Pu concentrations in the studied surface soil samples. While, the soil moisture content with factors related with the deposition process (annual precipitation rate, altitude, and topography) may be insignificant ones.

(1) LOI

The organic content can directly influence the retention of deposited plutonium in the soil. Although plutonium could also be associated to oxides and minerals in the soil, over 50% of plutonium in the soil was observed as organic matter associated form (Lee and Lee, 2001), and the association of plutonium to macromolecules can significantly reduce the migration of plutonium in the environment (Ni et al., 2018; Bu et al., 2014). Additionally, the higher organic content in soil is usually associated with higher vegetation coverage, which can limit the loss of plutonium in the atmosphere through suppressing the soil erosion (Zhang et al., 2021). The similar retention function of soil organic matter content on the concentration of fallout Pu in surface soils has also been reported in other locations (Lee and Lee, 2001; Komosa, 1999; Zhang et al., 2021).

(2) Human activities

Human activities of plowing, irrigation and road construction can heavily disturb the soil. $^{239,240}$Pu deposited in the surface soil is highly mixed during the plowing ($25-35$ cm) and road construction (>35 cm), causing a relatively homogeneous distribution of plutonium in this kind of soil profiles (Zhao et al., 2020; Zhang et al., 2021). Consequently, the plutonium concentrations in the surface soil were decreased in such disturbed soil. Irrigation promotes migration of plutonium from surface layer to the deep layer, and reduce the concentration of plutonium in the top layer as well. Therefore, the human activity was manifested as a key factor in the distribution of plutonium. Such influence has also been observed in Northern Xinjiang (Zhao et al., 2020), northeast China (Xu et al., 2013), the Loess Plateau (Zhang et al., 2019), South Korea (Lee and Lee, 2001).

(3) Soil moisture content

A linear correlation between soil moisture content and plutonium concentrations in the investigated soil was observed, but the multiple linear regression analysis does not show a significant correlation of soil moisture. This might result from two issues: (1) plutonium is not a mobile element in the environment, which could be highly associated with organic substance, oxides, and mineral fractions in the soil, and less present as the water soluble, exchangeable and carbonate fraction (Lee and Lee, 2001); (2) the soil moisture might only be an instantaneous parameter and closely related to the organic content. Besides, the soil characteristics and human activity, the soil moisture content is dependent on the precipitation rate, evaporation rate, and varies within time. While, soil organic content is independent of climate, but influenced by the vegetation coverage and input of organic matters (Xie et al., 1996). A significantly high correlation ($r = 0.78$) between soil moisture content and LOI was observed in the studied soil samples, indicating their close relation. For example, the mild climate and high precipitation always increase the soil moisture content and promote the vegetation coverage and therefore the content of the organic substance in soil. Therefore, only the more “stable” organic content was identified as a key factor in the multiple linear regression model.

(4) Annual precipitation rate

It was reported that plutonium in the atmosphere could be effectively removed to the surface by wet deposition, i.e. precipitation flushing (Lee and Lee, 2001; Hirose et al., 2004). However, the annual precipitation rate was not identified as a key factor for the plutonium level in surface soil. The comparable mean $^{239,240}$Pu concentrations were measured in surface soil collected from the humid region (0.359 mBq/g with annual precipitation rate> 800 mm), semi-humid region (0.382 mBq/g with annual precipitation rate of 400–800 mm) and semi-arid region (0.334 mBq/g with annual precipitation rate of 200–400 mm).

First of all, the plutonium concentration in atmosphere from the studied area ($27.5' – 36.4' N$) should be much less than that in the close-in region of tests sites, as well as in the higher altitude belt (40–50’ N) due to the global fallout origin and its conservative circulations characteristics (UNSCEAR, 2000). In most sites (>50%) of the studied area, the annual precipitation rates are less than 500 mm, and most precipitation in the studied occurs in short wet seasons (http://www.cma.gov.cn/). Therefore, the wet deposition of global fallout of the nuclear weapons tests is not effective, and therefore accounts for small contributions to the total deposition of plutonium. Additionally, the relatively low annual precipitation rate was not a key factor on the migration of plutonium that deposited and strongly associated in soil particles. As a result, the annual precipitation rate was not a key factor affecting the distributions of plutonium concentrations.

(5) Altitude and topography

The statistical analysis shows that the altitude and topography do not show significant effects on the plutonium concentrations in the surface soil. The resuspended plutonium during dust storm in troposphere has been observed to accumulate in the mountain front areas in Northern China (Zhang et al., 2021; Hirose et al., 2003). However, the relatively
low dust flux occurred in the Qinghai-Tibet Plateau compared to surrounding areas (Gong et al., 2012), the contribution of the re-suspended plutonium within dust in this region is therefore not significant. Other pollutants including black carbon, heavy metal, persistent organic pollutants emitted from neighboring industrial manufactures in South Asia and dispersed near the surface and in the troposphere have also been observed to preferentially deposit in the mountain front areas of the Qinghai-Tibet Plateau (Guo et al., 2020; Kang et al., 2019). But, due to the long distance and relatively low concentrations of plutonium in atmosphere from the studied area, such preferential deposit process seems not happen. The observed higher concentrations in mountainous areas should be attributed to the high higher LOI values (mean: 22%) and less disturbance from human activities.

In the sum, the key process that influence the spatial distribution of plutonium in surface soil from the Qinghai-Tibet Plateau is the retention instead of the deposition process, and the organic content and human activity are the two key parameters influencing the retention of plutonium in the soil. The seven highest values from 0.805 to 1.95 Bq/kg were mainly attributed to the well retention of deposited plutonium in the soil samples with high organic content and less human activities disturbances.

5. Conclusions

Based on $^{239}$Pu and $^{240}$Pu concentrations determined in the surface soil samples collected from the Qinghai-Tibet Plateau and discussion above, it can be concluded that (1) the $^{239,240}$Pu concentrations (0.0176–1.95 Bq/kg) and the $^{240}$Pu/$^{239}$Pu atomic ratios (0.146–0.225) in the surface soil in Qinghai-Tibet plateau indicate that the global fallout is the major source of plutonium in this region, and the plutonium level is too low to trigger radiation risk; (2) annual precipitation rate, altitude and topography of the site related to the deposition process of plutonium make less effect on the distribution of plutonium in the studied surface soil; (3) organic matter content in the soil and human activities have a significant effect on the distribution of plutonium, with high plutonium concentrations measured in the samples of higher soil organic content and less disturbance from human activities.

Credit author statement


Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.envpol.2022.119401.


