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Level, distribution and sources of plutonium in the northeast and north China

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ABSTRACT

Concentrations of ^{239}Pu and ^{240}Pu in 163 surface soil samples and five soil cores collected from the northeast and north China were analyzed using the radiochemical separation combined with inductively coupled plasma mass spectrometry measurement. The average ^{240}Pu/^{239}Pu atomic ratios (0.185 ± 0.018) for all surface soil samples indicated that the global fallout is the major source of plutonium in the studied region. The ^{239,240}Pu concentrations of the surface soil ranged from 0.002 mBq/g to 4.82 mBq/g, lying in the range of the reported results in the areas with similar latitude, except for a few samples. The distribution of ^{239,240}Pu in this region is controlled by the deposition of plutonium in the atmosphere and its preservation in the soil, which were affects by multi-factors such as topography, climate, utilization of the land and vegetation coverage. The analytical results could be used as the baseline data for the assessment of the impact of nuclear activities in the past and the future.

1. Introduction

Since the 1940s, artificial radionuclides, such as ^{238,239,240}Pu, ^{137}Cs and ^{131}I, have been released to the environment by nuclear weapons tests, nuclear accidents, nuclear power plants and nuclear fuel reprocessing facilities (UNSCEAR, 2000). For example, it has been reported that 165 TBq of ^{239,240}Pu has been released from the 133 atmospheric nuclear weapons tests at Semipalatinsk of the former Soviet Union during 1949–1962 (Kadyrzhanov et al., 2005). The released radioactive matter could be widely dispersed and deposited on the ground. A relatively high level of ^{129}I in the surface soil samples has been observed in the northeast of China compared to the south of China (Fan, 2013; Lu, 2018; Zhang et al., 2015).

The ^{240}Pu/^{239}Pu atomic ratio in the sample from different source are different. A ^{240}Pu/^{239}Pu atomic ratio of 0.178 ± 0.023 in the environmental samples only received global fallout of the atmospheric nuclear tests was observed. While, different ^{240}Pu/^{239}Pu atomic ratios were observed in the close-in deposition of nuclear weapons tests, a ^{240}Pu/^{239}Pu atomic ratio of 0.30–0.36 was determined in the close-in deposition in the Pacific Proving Grounds (American Pacific nuclear weapons tests sites). The ^{240}Pu/^{239}Pu atomic ratio of weapons-grade plutonium was reported to be 0.01–0.07, but much higher values of 0.23–0.67 were observed in the releases from nuclear power plants (Smith et al., 1995; Varga, 2007; Warneke, 2002; Yamamoto et al., 2004), and ^{240}Pu/^{239}Pu atomic ratios of 0.38–0.41 for releases from the Chernobyl accident and 0.32–0.33 for Fukushima accident were observed (Boulyga et al., 1997; Wendt et al., 1999; Zheng et al., 2013), respectively. Therefore, ^{240}Pu/^{239}Pu atomic ratio has been used as a fingerprint to identify the source of the associated radioactive substance in the environment. Moreover, based on the ^{240}Pu/^{239}Pu atomic ratio, the plutonium could also be used for the evaluation of the contribution of nuclear activity in the soil (Dong, 2010; Ni et al., 2018; Xing, 2015; Xu, 2014; Zhang and Hou, 2019; Zhang et al., 2019).

Although plutonium isotopes in some soil samples collected in a few

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locations in northeast and north China have been analyzed (Dong, 2010; Ni et al., 2018; Wang et al., 2013; Xing, 2015; Xu, 2014; Zhang and Hou, 2019), a systematic investigation on its distribution and sources have not yet reported. With an increased number of nuclear power plants in China, including those in the northeast and north China, their radiation impact on the local environment has become a high concern.

The distribution of ^{239,240}Pu in the environment is not only related to the sources, factors influence its dispersion (e.g., wind direction), but also related to topography and environmental factors. A relatively high ^{239,240}Pu levels were observed in mid- and high-latitude bands of the North Hemisphere (UNSCEAR, 2000; Zhang and Hou, 2019), which was because most atmospheric nuclear weapons tests were conducted in the mid- and high-altitude in the Hemisphere. A good correlation of ^{239,240}Pu concentration with soil properties (organic matter content, soil particle size) (Lee et al., 1997; Xu, 2014; Zhang et al., 2010) and vegetation coverage (Ni et al., 2018; Xu, 2014) have been observed.

This work aims to systematically investigate the level and distribution of plutonium isotopes (^{239}Pu and ^{240}Pu) in the soil of the northeast and north China to better understand the sources of plutonium in this region. The impacts of topography (latitude, mountain), climate (wind field, precipitation) and environment factors (anthropogenic disturbance, vegetation coverage, organic matter content) on its spatial distribution of plutonium isotopes will be also investigated.

2. Materials and methods

2.1. Samples

Surface soil samples (0–5 cm for 133 samples or 0–30 cm for 30 samples in farmland) and 5 soil cores were collected from the northeast and north China (as shown in Fig. 1) during 2013–2016. Soil cores were collected from 5 sites with different types of land, i.e., desert (C1), grassland (C2, C4) and farmland (C3, C5). The soil samples of 0–30 cm depth and 4 soil cores of 0–35 cm were collected using a stainless-steel soil core sampler of 7.0 cm in diameter and 35 cm in length. The soil layers in each 5.0 cm interval were sliced. One soil depth profile (C1) of 30 cm depth with 2.0 cm intervals were collected using a stainless steel spade. All surface soil samples were collected using a cylinder soil sampler of 8 cm in diameter and 5 cm in height. Soil at each site was sampled at 5 points in a square of 1 m length between each point at each site, the sliced samples from the same depth were mixed as one sample. The collected soil samples were sealed in plastic bags and transported to the laboratory for analysis.

After removal of big stones (>3 mm in diameter) and vegetation roots (>5 cm in length) manually, the soil was weighed and air-dried. The samples were further dried in the oven at 150 °C until constant weight. The dried soil was weighed, ground and sieved through 80-mesh sieve.

2.2. Analytical methods

A radiochemical separation coupled with ICP-MS/MS measurement was used for determination of low-level of ^{239}Pu and ^{240}Pu. The detailed analytical method has been reported elsewhere (Zhang and Hou, 2019), a brief description is given here: (1) the prepared soil samples were ashed at 450 °C overnight, and the loss of samples weight during ash was recorded as the organic matter content in soil; (2) the ashed sample was leached with *aqua regia* after spiking a known amount of ^{242}Pu as the chemical yield tracer; (3) plutonium was separated from matrix by iron hydroxides co-precipitation; (4) the plutonium were purified using an extraction chromatography with TEVA column; (5) the final solution containing plutonium in 0.5 mol/L HNO_3 was measured using ICP-MS/MS (Agilent 8800) with the NH_3–He as the reaction-collision gas for ^{239}Pu, ^{240}Pu and ^{242}Pu, the concentration of ^{239}Pu and ^{240}Pu in the samples was calculated based on the amount of ^{242}Pu spiked and the measured signal intensity of ^{239}Pu, ^{240}Pu and ^{242}Pu in consideration of

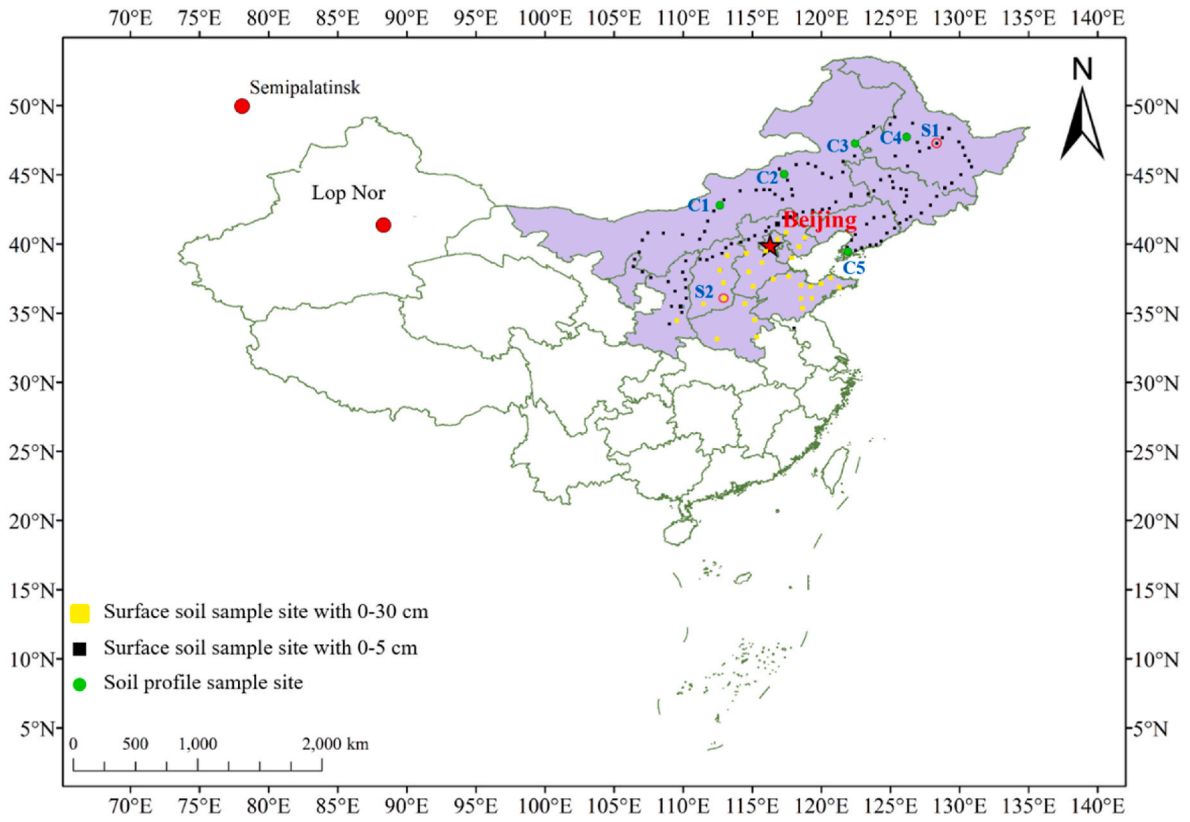

Fig. 1. Sampling sites of surface soil (black square for 0–5 cm depth sample, yellow square for 0–30 cm) and soil profile (green circle) in the Northeast China. (For interpretation of the references to colour in this figure legend, the reader is referred to the Web version of this article.)

the weight of the sample. The $^{240}Pu/^{239}Pu$ atomic ratio in the samples was directly measured by the signal intensity of these two isotopes by subtraction for the contribution of the procedure blank.

Certified reference materials soil (IAEA-327) and sediment (IAEA-385) and blank samples were analyzed using the same procedure as samples for quality control. The recoveries of plutonium monitored by the spiked ^{242}Pu tracer range from 76 % to 99 %. The analytical results of ^{239}Pu and ^{240}Pu in the certified reference materials are in good agreement with the certified values.

3. Results and discussion

3.1. Level and distribution of plutonium in the northeast and north China

The ^{239}Pu and ^{240}Pu level in all 163 surface soil samples are shown in Fig. 2, in which $^{239,240}Pu$ activity concentrations (^{239}Pu plus ^{240}Pu) were calculated for easy comparison with the reported data measured by alpha spectrometry. $^{239,240}Pu$ concentrations range from 0.002 mBq/g to 4.82 mBq/g, which are comparable to the reported values (0.002–0.880) in surface soil from the region without direct contamination (Dong, 2010; Wang et al., 2013; Xing, 2015; Xu, 2014; Zhang and Hou, 2019; Zhang et al., 2019), with a few exceptional high data.

As to the general geographical distribution, relatively higher $^{239,240}Pu$ concentrations were observed mainly in two areas, i.e., west of Greater Khingan Range and east of Changbai Mountain (Fig. 2), while relatively lower level plutonium was observed in the region between the Greater Khingan Range and Changbai Mountaint, as well as other areas in lower latitude including Shanxi (0.011–0.328 mBq/g), Henan (0.034–0.138 mBq/g), Southern Hebei (0.088–0.156 mBq/g) and Shandong (0.015–0.142 mBq/g).

This distribution generally agrees with the normal latitude distribution of global fallout radionuclides in north hemisphere (Kelley et al., 1999; Thakur et al., 2017; UNSCEAR, 2000), i.e., the concentration of deposited plutonium in the environment gradually increases with the latitude in 30–50 °N. Fig. 3 shows the distribution of $^{239,240}Pu$ concentration with the latitude. The average $^{239,240}Pu$ concentrations in each 5° latitude band are 0.131 ± 0.038 mBq/g in 30–35 °N, 0.107 ± 0.089 mBq/g in 35–40 °N, 0.289 ± 0.331 mBq/g in 40–45 °N and 0.540 ± 0.863 mBq/g in 45–50 °N. The lowest $^{239,240}Pu$ in the surface soil was observed in the area of the lower latitude of 35–40 °N, while the highest $^{239,240}Pu$ concentration average was observed in the area of the highest latitude of 45–50 °N.

The higher $^{239,240}Pu$ level in the mid- and high-latitude (40–50 °N) area is attributed to that the most atmospheric nuclear weapons tests were conducted in 40–50 °N, and the radioactive substances injected to the troposphere and stratosphere were deposited mainly in the similar latitude bands. The larger particles released into the troposphere mainly deposited in close-in area, while volatile substances and fine particles dispersed into the upper troposphere were dispersed to a large area. The debris released from a high yield weapon (such as thermonuclear devices) was mainly injected into the stratosphere and upper troposphere, which were mostly dispersed in a similar latitude band (Kelley et al., 1999; UNSCEAR, 2000).

The lower $^{239,240}Pu$ level in 35–40 °N compared to 30–35 °N might be attributed to artificial disturbance. The surface soil samples in 35–40 °N were mainly collected in farmland because this band is the most important farming area in China, and undisturbed soil is hardly found. The deposited plutonium in the surface layer soil was frequently mixed in the plowing layer with farming, and diluted by mixing with low plutonium deep soil. Thus, a relatively homogenous distribution of $^{239,240}Pu$ in the soil core of the upper 30 cm was observed in the soil core of C5 (see Fig. 4(e)).

The lowest $^{239,240}Pu$ concentration (0.002 mBq/g) was observed in sample S2 collected from cultivated farmland in Changzhi city in Shanxi province (37.2 °N, 112.9 °E). Besides its low latitude, artificial disturbance by agriculture activity which mixed or turn over the soil is another reason causing the decrease of plutonium concentration via diluting. On the other hand, the highest $^{239,240}Pu$ concentration (4.82 ± 0.14 mBq/g)

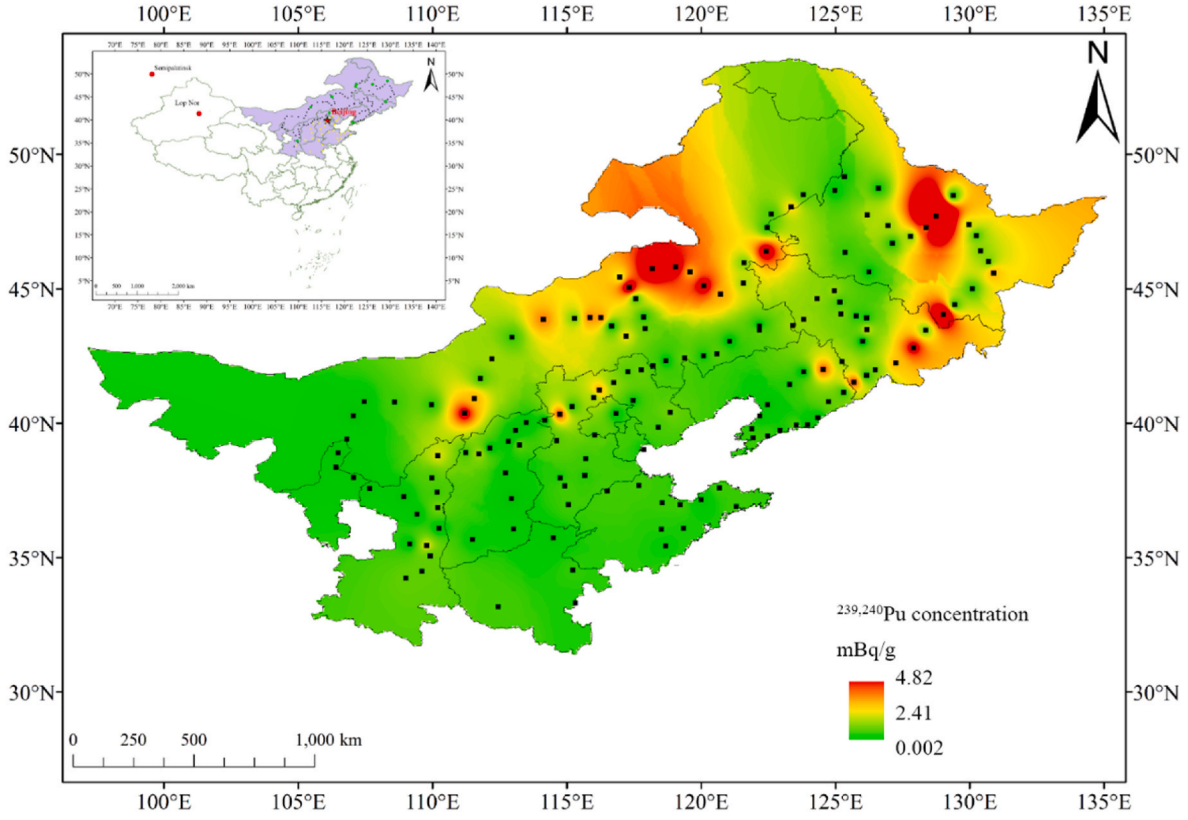

Fig. 2. Distribution of $^{239,240}Pu$ concentrations in the surface soil in the Northeast China.

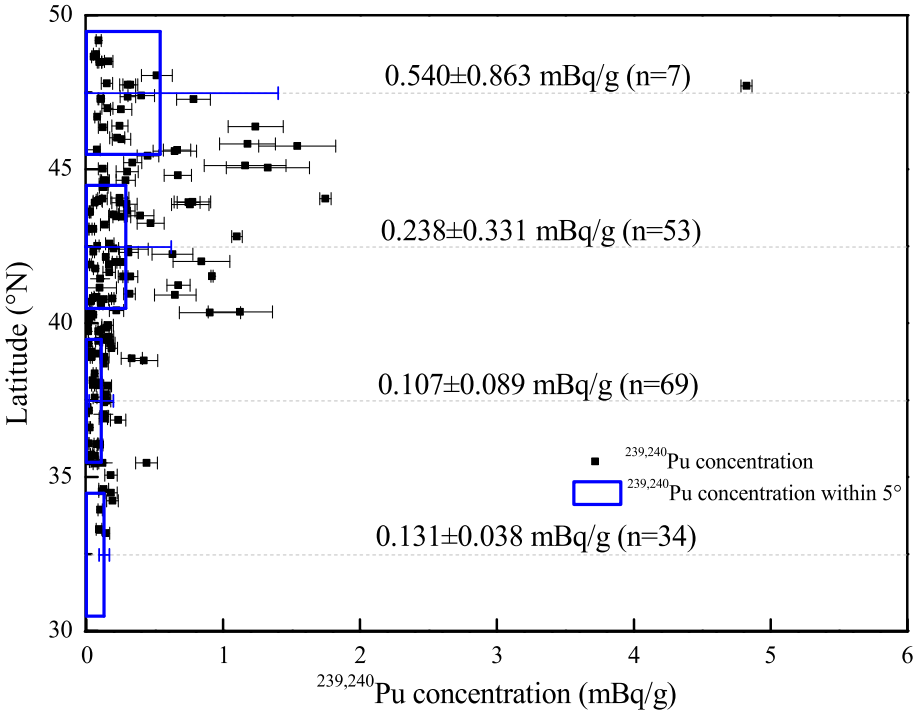

Fig. 3. Distribution of ^{239,240}Pu concentrations in the surface soil with latitude in 30–50 °N.

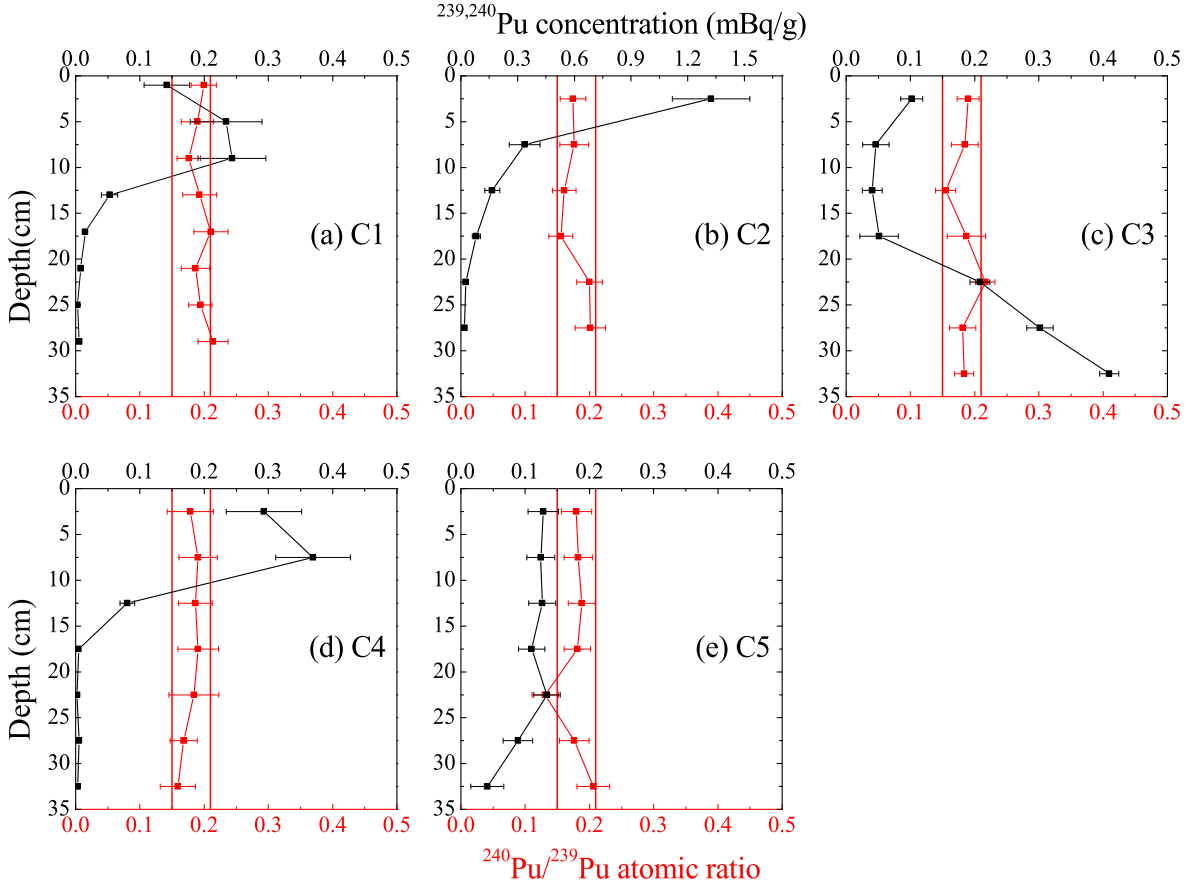

Fig. 4. Depth distribution of ^{239,240}Pu concentrations and ^{240}Pu/^{239}Pu atomic ratio in five soil profiles.

was observed for soil sample S1 collected from a forest at Yichun city in Heilongjiang province (47.7 °N, 128.7 °E). Besides the high deposition of this latitude band, a high amount (53 %) of organic substance and climate characteristics also contribute as discussed later.

3.2. Sources of plutonium isotopes in the northeast and north China

Distribution of $^{240}Pu/^{239}Pu$ atomic ratios in the surface soil from northeast and north China is presented in Fig. 5. The measured $^{240}Pu/^{239}Pu$ atomic ratios ranged from 0.141 ± 0.061 to 0.227 ± 0.057, and the relatively big variation of atomic ratio is mainly attributed to the high measurement uncertainty of low level samples, especially for ^{240}Pu which is much lower than ^{239}Pu in mass concentration. The $^{239,240}Pu$ concentrations with the lower or higher ratio are lower than 0.05 mBq/g, the higher measurement uncertainty (20–50 %) of ^{240}Pu were obtained in the ICP-MS/MS measurement, causing a big variation of atomic ratio. The average value of $^{240}Pu/^{239}Pu$ atomic ratios (0.185 ± 0.018) in the surface soil in the northeast and north China agree well with the reported value of global fallout from nuclear weapons tests (0.178 ± 0.023) (Koide et al., 1985; Krey et al., 1976), and significantly higher than that of weapons-grade plutonium and close-in deposition of nuclear weapons tests (0.01–0.07) (Chiappini et al., 1999; Hrnecek et al., 2005; Wolf et al., 1997; Yamamoto et al., 2004), meanwhile obviously lower than that of releases from nuclear power plants including Chernobyl accident and Fukushima accident (0.23–0.67) (Boulyga et al., 1997; Muramatsu et al., 2000; Zheng et al., 2013), indicating that the global fallout of the atmospheric nuclear weapons tests in 1945–1980 is the dominant source of plutonium in the northeast and north China.

In the Chernobyl accident in 1986 and Fukushima accident in 2011, the large amounts of radioactive substances was released to the environment. As a particle-associated element, plutonium released was mainly deposited in the surrounding areas of the accident sites. Plutonium from the Chernobyl accident was mainly observed in nearby areas in the Ukraine, Belorussia and Russia (Steinhauser et al., 2014; UNSCEAR, 2000), and the Fukushima-derived plutonium was observed and detected only in the adjacent (<20 km) of Fukushima Daiichi NPP (Zheng et al., 2013). The contribution from these accidents to the plutonium in the northeast and north China is negligible.

The sampling sites are far away from the nuclear weapons test sites, about 1200 km from the nearest site at Lop Nor, and more than 2000 km from the Semipalatinsk nuclear weapons tests site. Some radioactive substances had been released from the atmospheric nuclear weapons tests at Lop Nor during 1964–1980 and might be transported to and deposited in the downwind area of the westerlies in northern China. As their emission was much smaller compared to the global fallout (UNSCEAR, 2000), the contribution of Chinese nuclear tests is indistinguishable from the global fallout. $^{240}Pu/^{239}Pu$ atomic ratios similar to that of global fallout were observed in the surface soil samples and soil profiles from downwind areas in China (0.147–0.221) (Dong, 2010; Wang et al., 2013; Xing, 2015; Xu, 2014; Zhang et al., 2018; Zhang and Hou, 2019; Zhang et al., 2019; Zheng et al., 2009), and in sediment from Lake Shuangta (0.160–0.192) and Lake Sihailongwan (0.160–0.192) in China (Wu et al., 2010). These data indicate that the contribution of close-in deposition of China atmospheric nuclear weapons tests on the plutonium in the soil in the downwind area far away from nuclear activity areas is minor and can be neglected compared to the global fallout.

On the other hand, the $^{240}Pu/^{239}Pu$ atomic ratios in 16 surface soil samples from the adjacent of Hongyanhe nuclear power plant in Liaoning Province (0.157–0.211) fluctuate around the ratio of the global fallout, and agrees well with the reported values (0.162–0.213) for surface and soil profile samples in Liaoning province (Xu, 2014; Zhang et al., 2018). Therefore, the effect of nuclear power plants on environmental radioactivity can be neglected.

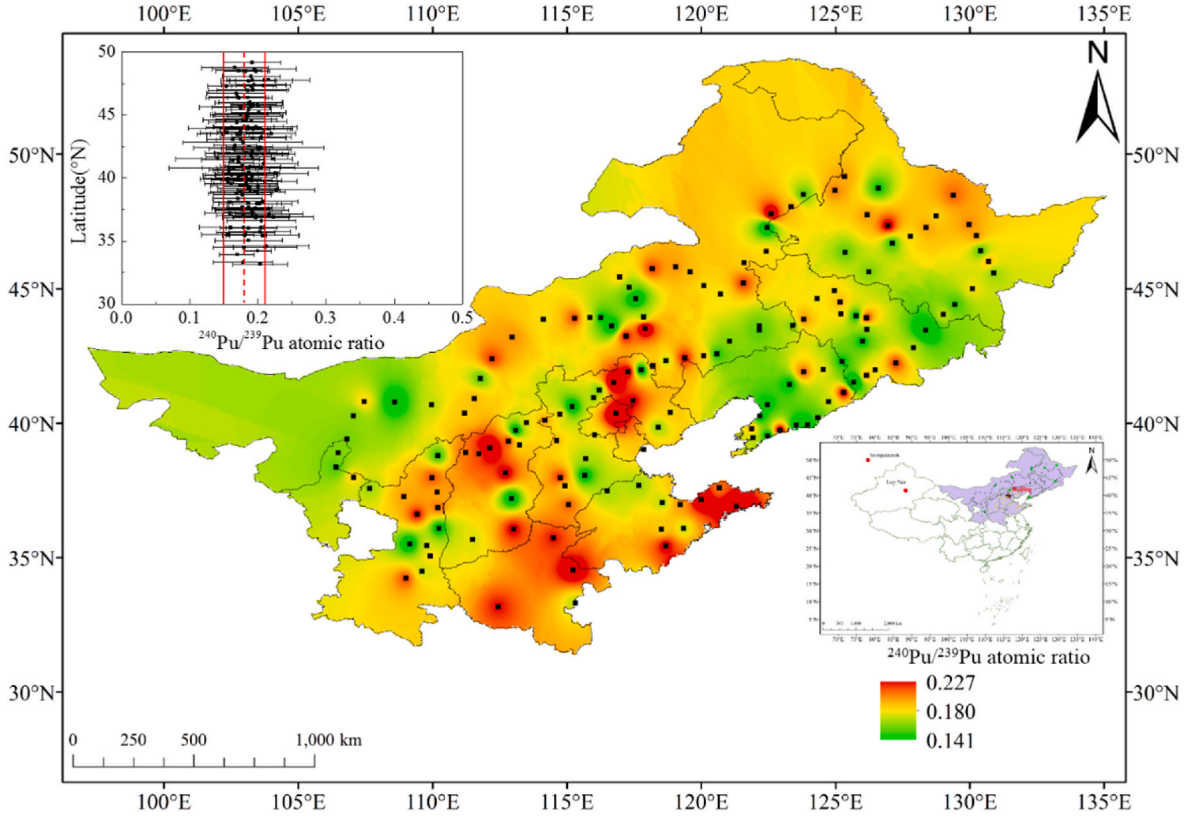

Fig. 5. Spatial distribution of $^{240}Pu/^{239}Pu$ atomic ratios in the surface soil in the Northeast China.

3.3. Effect of environmental parameters on the distribution of plutonium in the surface soil in the northeast and north China

3.3.1. Effect of vegetation coverage and soil organic substance content

In the investigated area, high ^{239,240}Pu concentrations (>1 mBq/g) were observed in surface soil from the undistributed forest and grassland with high organic matter content (>10 %) and vegetation coverage compared to other types of soil with the relatively lower organic matter content (Table 1, Fig. 2). The highest ^{239,240}Pu concentration (4.82 mBq/g) was observed in a forest soil with high organic content up to 53 %, while the plutonium concentrations in the low organic content and less vegetation coverage sandy soil were the lowest. The depth distributions of ^{239,240}Pu concentrations were also different for different types of soil. The maximum plutonium was observed in the surface (0–5 cm) soil in grassland soil (C2), which is one order of magnitude higher than the maximum value observed in the sub-surface (8–10 cm) in sandy soil (C1), meanwhile, the organic substance contents in grassland soil profile C2 (4.4–5.6 %) were also significantly higher than that in the sandy soil profile C1 (1.3–3.8 %). And a significant positive correlation ($R^2 = 0.55$) between ^{239,240}Pu concentration and the organic substance content in soil (see Fig. 6, $p < 0.01$) was observed. Similar results were also observed in other locations (Lee and Lee, 2001; Lee et al., 1997; Xu, 2014; Yamamoto et al., 1981).

Fractionation analysis of soil and sediment has shown that ^{239,240}Pu is mainly associated with organic substance due to the high adsorption of plutonium onto organic matter, which inhibits the migration of plutonium. On the other hand, the higher organic substance content is associated with higher vegetation coverage, which can effectively suppress soil erosion (Fu et al., 2009; Li et al., 2009; Zhang et al., 2019). Consequently, the deposited plutonium is effectively reserved in the surface soil.

3.3.2. Effect of topography and climate

The spatial distribution of ^{239,240}Pu in the surface soil in northeast and north China is quite inhomogeneous (Fig. 7): relatively higher ^{239,240}Pu concentration was observed in the north of Yin Mountain, northwest of Greater Khingan Range, and east of Changbai Mountain, compared to other areas. This might be attributed to the influence of the topography (mountains, etc.) and climate (wind direction, precipitation rate, etc.) on the dispersion and deposition of particle-associated plutonium in this region.

Besides direct fallout after each atmospheric nuclear weapons tests, resuspension and further dispersion of the deposited radioactive particles from a nearby area of the nuclear weapons tests sites to the sampling area might be another source of plutonium in the investigated region, especially through sandstorms which occur frequently in winter and spring in the north and northwest China (Qian et al., 2002). It has been observed that the plutonium-bearing particles deposited from global fallout could be resuspended, transported and deposited in other areas of East Asia (Charney, 1947; Holton, 1973; Liu et al., 2004).

Table 1
^{239,240}Pu concentration and organic substance content in the surface soil of different types of land.

Type of soil	Sampling site	^{239,240}Pu concentration (mBq/g)			Organic substance content (%)	
		Max	Min	Average	Max	Min
Close to river	8	0.198	0.031	0.091	9.7	0.8
Farmland	79	0.916	0.002	0.173	16	0.6
Sandy	6	0.228	0.003	0.170	5.2	1.1
Grassland[a]	24	0.465	0.005	0.193	16	1.5
Grassland[b]	12	1.538	0.446	0.902	18	3.7
Forest[a]	28	0.334	0.011	0.156	15	1.6
Forest[b]	6	4.824	0.643	1.700	53	8.7

Note: a-the sampling site with the disturbance; b-the undisturbed sampling site.

Geographic and geomorphic conditions have an important effect on the deposition of radionuclides from atmosphere to the ground, and therefore the distribution of plutonium in the northeast and north China. The westerlies are the dominant wind in North China with East Asian winter monsoon occurring in the winter. The westerlies can transport particles from source areas with high deposition of nuclear weapons tests derived radioactive particles to the study areas and even South Korea and Japan (Choi et al., 2006; Dong et al., 2017; Hirose et al., 2010). The Greater Khingan Range located in the northeast China extending 1200 km from north to south with an average altitude of 1400 m, the Yin mountains located in the north China extending 1000 km from west to south with an altitude of 2000 m, the radioactive particles carried by air masses at lower altitude transported from west to east by westerlies and from north to south by East Asian Monsoon could be deposited on the front of the mountains causing a relatively high inventory of plutonium in the west of the Yin Mountain and the northwest of Greater Khingan Range (Fig. 2). It was observed that the inventory of plutonium at site C2 (175 Bq/m^2) in Xilingol League in Inner Mongolia is two times higher than that at site C4 (83.2 Bq/m^2) in Qiqihar in Heilongjiang province. Besides, the sloping terrain of the Greater Khingan Range has an obviously increased deposition of atmospheric particles carried by westerlies at the low-altitude, causing the deposition and accumulation of particles associated plutonium in the front of the Greater Khingan Range when the air mass come from the northwest and northern regions, such as Mongolia (Hu et al., 2016; Marx et al., 2008; Peckham and Wicker, 2000; Smith, 2010). When plutonium-bearing particles deposited on the land with higher vegetation coverage and organic substance content, the plutonium is easily reserved in the deposition site, resulting in a relatively higher ^{239,240}Pu concentration and inventory.

For the high-altitude air mass (more than 5000 m), the influence of mountain on the deposition of plutonium will be little because these mountains are lower than 3000 m (the highest altitude in the Greater Khingan Range, Yin Mountain and Changbai mountain are 1400 m, 2000 m, and 2691 m, respectively). However, high ^{239,240}Pu concentrations were also observed in the east of Changbai Mountain (Fig. 7), and the highest ^{239,240}Pu concentration (4.82 mBq/g) was observed in this area. The east area of Changbai mountain is located in the rear of the East Asian Trough which occurs in winter and spring. The rear of the East Asian Trough (see Fig. 8), which is in the Sea of Okhotsk, is helpful for the deposition of air mass carrying the atmospheric particles at high-altitude. When the East Asian Trough occurs, a large amount of high-altitude atmospheric particles carried by the westerlies and the winter monsoon are deposited (Charney, 1947; Holton, 1973; Liu et al., 2004).

The concentrations of plutonium in surface soil samples from the area of the band of 40–50 °N increase from the coast area to Inner Mongolia, in correspondence with the decrement of precipitation from >1000 mm/yr to <200 mm/yr (Weather China, 2021) (see Fig. 7). This seems controversial as great precipitation usually enhances the global fallout and may be interpreted with secondary migration and flourishing of particle-associated plutonium. High precipitation rate may decrease the plutonium concentration in the soil through soil erosion and vertical migration of plutonium. Different depth distributions of ^{239,240}Pu concentrations were observed in soil profiles collected at C2 and C4 both of grassland (Fig. 5(b) and (d)). The maximum plutonium concentration was observed in surface soil (0–5 cm) for C2 compared to the sub-surface soil (5–10 cm) for C4. The annual precipitation rate in C2 area (323 mm/yr) is lower than that in C4 area (precipitation: 500 mm/yr). This means high precipitation enhances the downward migration. However, the downward migration is primarily decided by the soil properties. For example, the maximum plutonium concentration in sample C1 profile, collected from the sandy soil and with lower precipitation rate (180 mm/yr), occurred in a deeper layer (8–10 cm).

3.3.3. Effect of artificial disturbance

Relative low concentrations of plutonium (0.015–0.215 mBq/g)

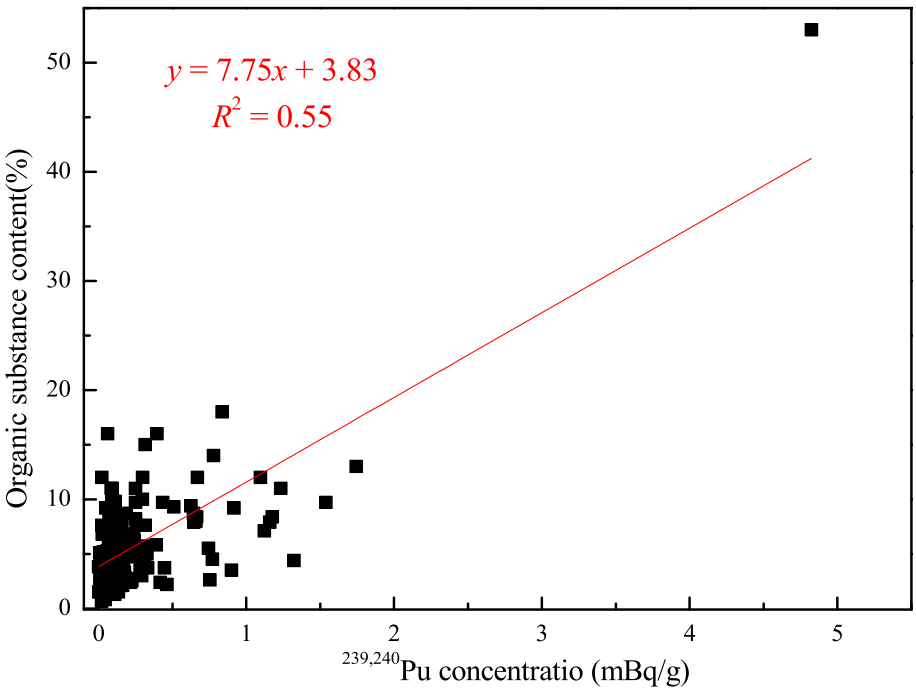

Fig. 6. The correlation between organic matter contents and ^{239,240}Pu concentrations in the surface soil.

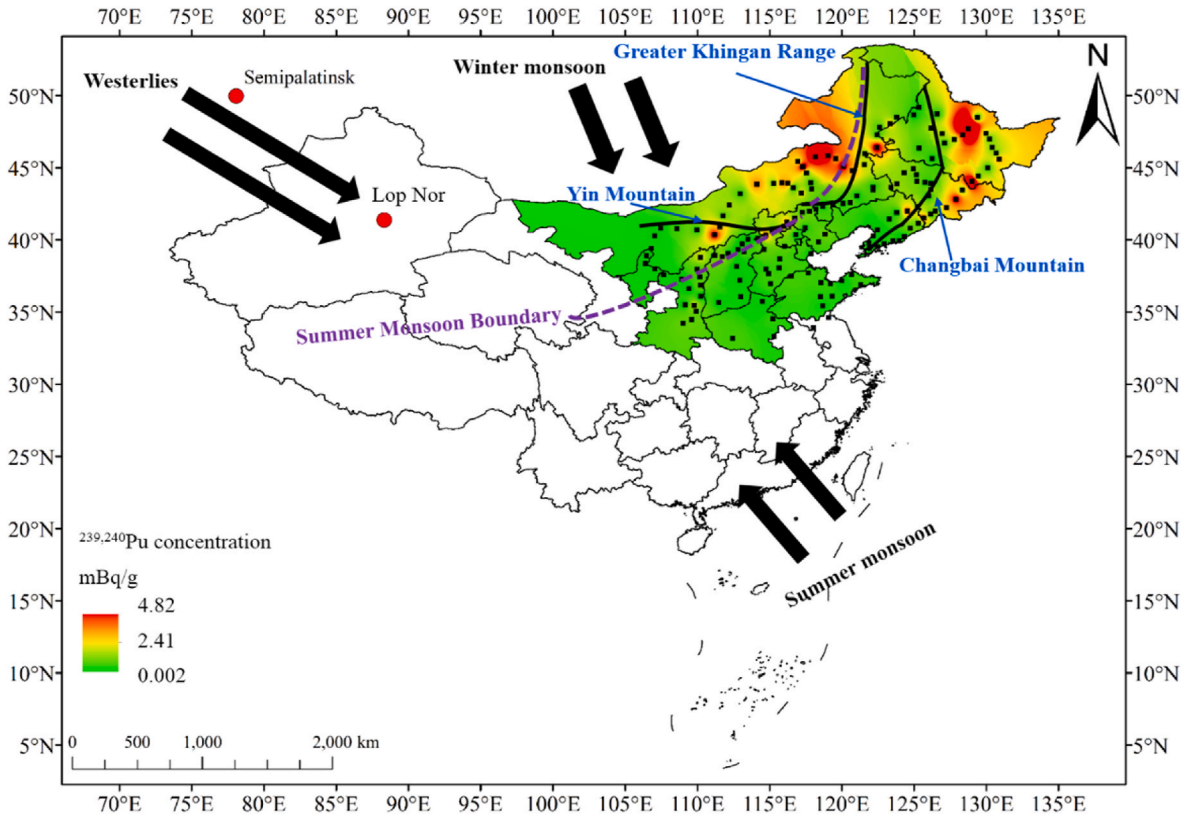

Fig. 7. Influence of topography and climate on distribution of ^{239,240}Pu concentration.

were observed in the surface soil from Shanxi, Shandong, Henan provinces and southern Hebei, compared to the other areas. This is mainly attributed to the artificial disturbance of the sampling site because of the high agriculture activities in these areas (Fig. 2). ^{239,240}Pu concentrations in surface soil in undisturbed sampling sites were always higher than those in sites with artificial disturbance (Table 1).

The depth distribution of ^{239,240}Pu concentrations in soil profile collected from the farmland (C5, Fig. 5(e)) shows a relative homogeneous level of plutonium in the upper 25 cm soil (plowing layer) compared to the exponentially decreasing profiles for most of the undisturbed site as shown in Fig. 4(b) (at site C2). While, the highest ^{239,240}Pu concentration was observed in the deep soil (25–35 cm) at the

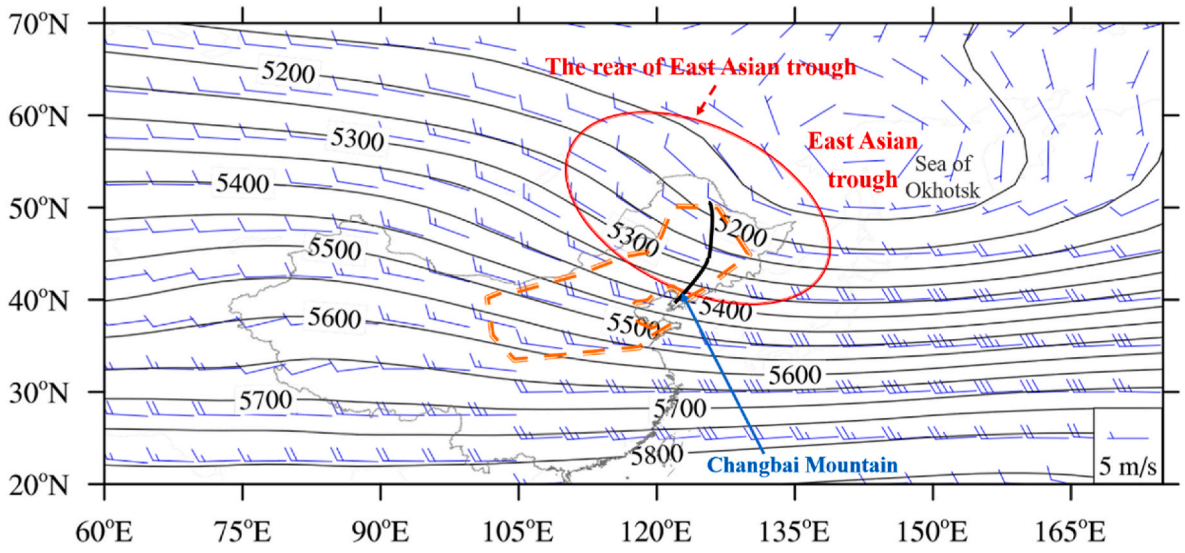

Fig. 8. Geopotential height and wind fields above 5000 m altitude in the winter of 1950–1980 (Charney, 1947).

disturbed sampling sites (e.g., Fig. 4(c)). This might be attributed to soil turnover by human activity. As a consequence, the ^{239,240}Pu concentration in the surface layer (0–5 cm) for disturbed soil in farmland is much lower than that for undisturbed soils (Fig. 4 and Table 1).

4. Conclusions

Based on the above results and discussion, it can be concluded: 1) the global fallout of atmospheric nuclear weapons tests is the dominant source of plutonium in the northeast and north China based on the narrow ranges of ^{240}Pu/^{239}Pu atomic ratios (0.141–0.228 with an average of 0.185 ± 0.017). 2) The ^{239,240}Pu concentrations (0.002–4.82 mBq/g) vary largely in the northeast and north China, which is related to the different deposition and fixation processes of plutonium in the soil in the study areas. 3) The organic substance content and vegetation coverage have a significant effect on the reservation of the deposited plutonium. 4) Mountains, westerlies and the East Asian Trough play an important role in the deposition of plutonium in this region. 5) Precipitation could cause decrease of the plutonium level in surface soil by promoting the migration of plutonium in the soil and the soil erosion. This paper reported the first set of systematic investigation of plutonium in the surface soil in the northeast and north China, which can act as the baseline values for the evaluation of the impact of nuclear activities in the last century as well as in the future.

Author contributions

The manuscript was written through contributions of all authors. Weichao Zhang implemented the samples analysis, Xiaolin Hou designed the research and supervised the reseaerch. Weichao Zhang, Yanyun Wang, Shan Xing and Ning Chen contributed to the sampling and ICP-MS measurement. Weichao Zhang, Xiaolin Hou, Haitao Zhang and Haijun Dang drafted and revised the manuscript. All authors have given approval to the final version of the manuscript.

Notes

The authors declare no competing financial interest.

Credit author statement

Zhang WC: Methodology, investigation, Writing- Original draft preparation, Visualization; Hou XL: Conceptualization, Writing –Review & Editing, supervision, project management, Funding acquisition; Zhang HT and Dang HJ: Writing –Review & Editing, supervision, Wang YY, Xing S and Chen N: Sampling and measurement, Writing -Review & Editing.

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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