Plutonium isotopes in Northern Xinjiang, China
Level, distribution, sources and their contributions

Zhao, Xue; Qiao, Jixin; Hou, Xiaolin

Published in:
Environmental Pollution

Link to article, DOI:
10.1016/j.envpol.2020.114929

Publication date:
2020

Document Version
Peer reviewed version

Citation (APA):

General rights
Copyright and moral rights for the publications made accessible in the public portal are retained by the authors and/or other copyright owners and it is a condition of accessing publications that users recognise and abide by the legal requirements associated with these rights.

- Users may download and print one copy of any publication from the public portal for the purpose of private study or research.
- You may not further distribute the material or use it for any profit-making activity or commercial gain
- You may freely distribute the URL identifying the publication in the public portal

If you believe that this document breaches copyright please contact us providing details, and we will remove access to the work immediately and investigate your claim.
Plutonium isotopes in Northern Xinjiang, China: Level, distribution, sources and their contributions

Xue Zhao a,b, Jixin Qiao a, Xiaolin Hou a,b,c,d *

a. Technical University of Denmark, Department of Environmental Engineering, Risø Campus, Roskilde 4000, Denmark
b. State Key Laboratory of Loess and Quaternary Geology, Xi'an AMS Center, Institute of Earth Environment, Chinese Academy of Sciences, Shaanxi Key Laboratory of Accelerator Mass Spectrometry Technology and Application, Xi'an 710061, P. R. China
c. CAS center of Excellence in Quaternary Science and Global Change, Xi'an 710061, P. R. China
d. Open Studio for Oceanic-Continental Climate and Environment Changes, Pilot National Laboratory for Marine Science and Technology (Qingdao), Qingdao 266100, P. R. China

ABSTRACT: Plutonium in the environment has drawn significant attentions due to its radiotoxicity in high concentration and source term linked with nuclear accidents and contamination. The isotopic ratio of plutonium is source dependent and can be used as a fingerprint to discriminate the sources of radioactive contaminant. 239Pu, 240Pu and 137Cs in surface soil and soil cores collected from Northern Xinjiang were determined in this work. The concentrations of 239,240Pu and 137Cs are in the range of 0.06-1.20 Bq kg⁻¹, and <1.0-31.4 Bq kg⁻¹ (decay corrected to Sep. 2017), respectively, falling in the

* Corresponding author: E-mail address: xiho@dtu.dk (X. Hou).
ranges of global fallout in this latitude zone. The $^{240}\text{Pu}/^{239}\text{Pu}$ atomic ratios of 0.118-0.209 and $^{239,240}\text{Pu}/^{137}\text{Cs}$ activity ratios of 0.039-0.215 were measured. Among the investigated sites, distinctly lower $^{240}\text{Pu}/^{239}\text{Pu}$ atomic ratios of 0.118-0.133 and higher $^{239,240}\text{Pu}/^{137}\text{Cs}$ activity ratios of 0.065-0.215 compared to the global fallout values were observed in the northwest part, indicating a significant contribution from other sources besides the global fallout. This extra source is mainly attributed to the releases of atmospheric nuclear weapons testing at Semipalatinsk Nuclear Test Site, which was transported by the west and northwest wind through the river valley among mountains in this region. This contribution is estimated to account for 28-43% of the global fallout in the northwest part of Northern Xinjiang. The contribution from the Chinese atmospheric nuclear weapons testing to this region is negligible due to the lack of appropriate wind direction to transport the radioactive releases to this region.

Keywords: $^{239,240}\text{Pu}$; $^{137}\text{Cs}$; radioactive pollution; Nuclear weapons testing; Northern Xinjiang

1. Introduction

Anthropogenic plutonium in the environment mainly originated from the global fallout of the atmospheric nuclear weapons tests in 1950-1960s, which injected large amount of radioactive substances including plutonium into the stratosphere, followed by a mixing process and deposited all over the world, but dominated in the North Hemisphere. Meanwhile, the radioactive substances released to the troposphere originated mainly from small yield nuclear weapons tests were deposited in the local
area of the nuclear weapons testing sites, and regional areas of some hundreds
kilometers from nuclear weapons testing sites. Spent nuclear fuel reprocessing plants
(NFRPs), nuclear accidents and nuclear facilities (e.g. nuclear power plants and nuclear
research facilities) also released some amount of plutonium to the environment, which
was confined mostly to the proximity of the sites due to the non-volatility of plutonium
(Alewell et al., 2017; Jiménez-Ramos et al., 2006). The released plutonium isotopes
have drawn significant attentions due to the high radiotoxicity of plutonium in high
concentration, especially the $\alpha$ emitting isotopes of $^{239}\text{Pu}$ ($T_{1/2}=24,110$ yr) and $^{240}\text{Pu}$
($T_{1/2}=6561$ yr). In addition, the fingerprint information carried by $^{240}\text{Pu}/^{239}\text{Pu}$ atomic
ratios can be used for identification of source terms and estimation of the contribution
of different sources. A lot of works have been conducted on the determination of
$^{239,240}\text{Pu}$ in soil samples, one of the most important sinks of plutonium (Francis and
Dodge, 2015), for investigation of the regional source terms of radioactive
contaminants, the environmental radioactive levels and transport pathways of the
radioactive substances (Yamamoto et al., 2004; Xu et al., 2013; Zhang and Hou, 2019a;
Jaegler et al., 2018; Turner et al., 2003; Olivier et al., 2004; Warneke et al., 2002).
 Northern Xinjiang might be affected by the two nearby nuclear weapons testing
sites, i.e. Semipalatinsk Nuclear Test Site (SNTS) in the northwest (600-1300 km), and
Lop Nor nuclear weapons testing site in the southeast (400-800 km). However, the
levels, distribution, sources and transport pathways of radioactive pollution in this
region are still unclear. Low $^{240}\text{Pu}/^{239}\text{Pu}$ atomic ratios (0.024-0.125) have been
measured in soil samples collected from the surrounding areas of the SNTS, indicating
the direct contamination from the former nuclear weapons tests (Yamamoto et al., 1999).

According to the investigation of \(^{239,240}\text{Pu}\) concentrations and \(^{240}\text{Pu}/^{239}\text{Pu}\) atomic ratios in soil and lake sediment samples from the adjacent Gansu Province, the regional deposition of plutonium from Lop Nor test site was clearly pointed by the high inventories up to 546 Bq m\(^{-2}\) and distinctly lower \(^{240}\text{Pu}/^{239}\text{Pu}\) atomic ratios (0.059) compared to the global fallout value (0.180) (Xing, 2015; Wu et al., 2010; Bu et al., 2015; Kelley et al., 1999). A few investigations of plutonium in nearby areas of Northern Xinjiang in early times have been implemented, and comparable levels of total alpha activities to global fallout were measured in soil samples from Central Xinjiang (Zhang et al., 1988b), but the plutonium isotope compositions and source terms were not measured due to the limitation of measurement techniques.

This work aims to investigate the level, distribution, source and transport pathway of radioactive pollution in Northern Xinjiang by determination of \(^{239}\text{Pu}\), \(^{240}\text{Pu}\) and \(^{137}\text{Cs}\) in surface soil and soil core samples collected in this region. Meanwhile the relative contribution of different sources to the radioactivity in this region will be also investigated based on the isotopic ratio fingerprints.

2. Materials and Methods

2.1 Studied area and sampling

The investigated area is located in Northern Xinjiang (43 °- 49 °N, 80 °- 96 °E) (Fig. 1), the hinterland of the Eurasian continent, which is dominated by the temperate continental arid and semi-arid climate. The Altai Mountains (47-53 ° N, 84-92 ° E, lie
from the northwest to southeast with elevations of 1000-3000 m) is in the north, the
Tarbagatai Mountains in the west, the Tianshan Mountains (lie from the west to east
with elevations of 1500-4000 m) in the south, and the Juggar Basin in the middle
hatched with the Gurbantunggut Desert. The temperature (mean annual temperature:
2.5-10 °C) and precipitation (mean annual precipitation: 20-600 mm) varies a lot in this
area due to the varied topography, such as intermountain basins, mountain areas, bolson
plain, etc. (Guo et al., 2017; Guli et al., 2015).

Fig. 1 Satellite image of the investigated area emphasized on Altai Mountains in the
north, Tarbagatai Mountains in the west, Tianshan Mountain in the south (yellow
triangles in lines), and Gurbantunggut Desert in the middle; sampling sites:
surface soil (black circle), soil core (blue circle); possible sources: red triangle;
Westerlies (blue arrow).
Surface soil samples (0-5 cm) were collected at 27 sites and soil cores (0-25 cm) were collected at 7 sites in Northern Xinjiang (except the Gurbantunggut Desert) in Sep. 2017 (Fig. 1). They were mainly collected from no artificial disturbed sites, such as in pastureland, desert steppe, grassland and Gobi Desert. Soil samples were also collected from the sites near roads and from cultivated lands, which were disturbed by human activities in some extend. The sampling sites correspond to different environmental and climate conditions in this region, including precipitation rates, vegetation coverages, soil types, human activities, etc. The soil cores of XJ-01, 02, 03, 05 and 07 were collected using a stainless-steel soil core sampler of 7 cm in diameter and 30 cm in height and sliced into 5 cm intervals. 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. At each sampling site, the samples from 3 points in a triangle of 1 m distance were collected and mixed as one. Two soil profiles of XJ-04 and XJ-06 were sampled from a hand-made square soil pit in 1 cm intervals. The collected soil samples were sealed in plastic bags and transported to the laboratory for analysis. The soil samples were 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.

The details of soil core samples were summarized in Table 1, wherein the vegetation coverage rate (the ratio of area with vegetation to the whole area in the viewing range) was estimated during sampling, bulk density was calculated using weight of the dried sample and volume, moisture content was calculated based on the
mass differences before and after drying, loss of ignition (LOI) was calculated based
on the mass differences before and after ashing in a muffle furnace at 450 °C for 12 h.

The sampling information for surface soil samples are listed in Table 1S in the
Supplementary Information.

| Table 1 Information of samples of 7 soil cores collected in Northern Xinjiang, China |
|---|---|---|---|---|---|---|---|
| Sample | Sampling site | Latitude /°N | Longitude /°E | Bulk density / g cm⁻³ | LOI /% | Average moisture content /% | Vegetation Coverage /% | Depth /cm |
| XJ-01 | Lakeshore | 92.82 | 43.61 | 0.72-1.07 | 7.65 | 19.2 | 60 | 20 |
| XJ-02 | Road | 85.96 | 44.28 | 0.85-1.22 | 3.83 | 2.20 | 30 | 25 |
| XJ-03 | Road | 87.04 | 47.71 | 1.78-2.51 | 2.45 | 8.74 | 30 | 25 |
| XJ-04 | Pastureland | 88.29 | 44.10 | 1.29-1.63 | 2.85 | 3.71 | 40 | 20 |
| XJ-05 | Pastureland | 83.42 | 43.53 | 0.64-0.77 | 9.30 | 10.7 | 100 | 25 |
| XJ-06 | Grass cluster | 83.95 | 44.67 | 2.49-3.07 | 5.58 | 1.68 | 50 | 20 |
| XJ-07 | Gobi | 90.30 | 43.86 | 1.45-1.67 | 4.98 | 3.31 | 20 | 20 |

2.2 Analysis of soil samples for isotopes of plutonium and ¹³⁷Cs

The activities of ¹³⁷Cs in the soil samples (50-100 g) was directly measured using
high-purity germanium (HPGe) γ -spectrometers with counting efficiencies of 25-40%
at the Technical University of Denmark (DTU), and the 661 keV gamma line was used
to measure the activity of ¹³⁷Cs. The counting time for each sample was around 80,000
seconds to reach a detection limit of 1 Bq kg⁻¹. The analytical uncertainty was better
than 10% depending on the ¹³⁷Cs level in the sample. The γ spectrometers were
calibrated for counting efficiency at different geometry using a mixed standard source,
the self-absorption and sum coincidence was corrected for each sample. All data of
¹³⁷Cs was decay-corrected to the sampling date of 10th Sep. 2017.
The plutonium isotopes in the soil samples were determined using the modified method from Qiao et al., (2009). In brief, an aliquot of 5-15 g soil was weighed into a beaker and ashed in a muffle furnace at 450 °C for 12 h to decompose organic matters in the sample, the ashed sample was weighed and loss of ignition (LOI) value was calculated (Table 1). The $^{242}$Pu of 5 mBq was spiked as a yield tracer to monitor the chemical yield of plutonium in the chemical separation procedure. *Aqua regia* of 50-150 ml was added to the beaker, and the sample was digested on a hotplate at 150 °C for 30 min and then 200 °C for 2 h. After filtration, ammonia was added to the leachate to adjust the pH to 8-9, plutonium was co-precipitated with Fe(OH)$_3$. The precipitate was dissolved with 5 ml of conc. HCl, K$_2$S$_2$O$_5$ was added to reduce all plutonium to Pu$^{3+}$, and then ammonia was added to pH 9-10. The precipitate was separated and dissolved with 3-5 ml of conc. HCl, and conc. HNO$_3$ was added to oxidize Pu$^{3+}$ to Pu$^{4+}$. The sample was adjusted to 3.0 M HNO$_3$ and loaded to a 2 ml TEVA column. The column was rinsed with 80 ml of 1 M HNO$_3$ and 40 ml of 6 M HCl. The plutonium on the column was finally eluted with 20 ml of 0.1 M of NH$_2$OH·HCl in 2 M of HCl solution, thereby reducing it to Pu$^{3+}$. The eluate was evaporated to dryness on a hot-plate at 200 °C, 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$_3$ for the measurement of plutonium isotopes.

The measurement of plutonium isotopes was conducted using a triple quadrupole ICP-MS (Agilent 8800, Agilent, Japan). The separated plutonium solution was transferred to a 10 ml tube, 50 μL of 100 μg L$^{-1}$ In(III) was added as
internal standard, and the sample was prepared in 3.0 ml of 0.5 M HNO₃ solution. The signals of $^{239}$Pu, $^{240}$Pu and $^{242}$Pu were measured by using ICP-MS equipped with and Xs-skimmer cone under hot plasma conditions, the single MS mode was employed in this measurement because of less uranium in the prepared samples. A high efficiency sample introduction system, Apex Q (Elemental Scientific Inc, Omaha, NE, USA) was used to introduce sample solution to the ICP-MS. With this system, the counting efficiency was improved by a factor of 6-8 through enhancing the sample introduction efficiency. The measurement sensitivity of $^{239}$Pu reached to 1600 cps ppt⁻¹. The detection limits were estimated to be 0.02-0.04 fg ml⁻¹ for $^{239}$Pu and $^{240}$Pu 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 1-2 orders magnitudes lower than that in samples. The concentrations of $^{239}$Pu and $^{240}$Pu in the sample solution were calculated based on the measured signal intensities of $^{239}$Pu, $^{240}$Pu and $^{242}$Pu, and the amount of $^{242}$Pu spiked to the samples before chemical separation. The isotopic fractionation of plutonium during the chemical separation and ICP-MS (<0.2%) is normally much lower than the analytical uncertainty (>2%), and therefore was not considered for correction. The detailed analytical method is presented in the Supplementary Material.
3 Result and interpretations

3.1 Plutonium isotopes and $^{137}$Cs in surface soil samples

The concentrations of $^{239,240}\text{Pu}$ (sum of $^{239}\text{Pu}$ and $^{240}\text{Pu}$) and $^{137}$Cs in 27 surface soil samples are presented in Table S1. A large variation of $^{239,240}\text{Pu}$ concentrations (0.06-1.20 Bq kg$^{-1}$) in these surface soil samples were observed, but falling into the range of the reported $^{239,240}\text{Pu}$ concentrations in surface soils from background areas (0.37-3.7 Bq kg$^{-1}$) (ATSDR, 2010). They are also similar to the reported $^{239,240}\text{Pu}$ concentrations of 0.36-0.68 Bq kg$^{-1}$ at the sites of 42 °N, 86 °E (Zhang et al., 1988b), 0.01-0.18 Bq kg$^{-1}$ at 41 °N, 84 °E, and 0.07-0.83 Bq kg$^{-1}$ at 37 °N, 82 °E (Dong et al., 2010a) in central Xinjiang.

The $^{239,240}\text{Pu}$ concentrations in the surface soil vary with the sampling environment and locations. Low $^{239,240}\text{Pu}$ concentrations of 0.06-0.24 Bq kg$^{-1}$ (n=6) were observed in samples collected near roads or from cultivated lands (S113, 114, 115, 116, 140 and 144). Higher $^{239,240}\text{Pu}$ concentrations of 0.30-0.67 Bq kg$^{-1}$ (n=10) were observed in samples collected from undisturbed areas with minor or non-vegetation coverage, such as Gobi and Steppe (S101, 103, 106, 109, 112, 147, 121, 122, 150 and 129). While the highest $^{239,240}\text{Pu}$ concentrations of 0.77-1.20 Bq kg$^{-1}$ (n=11) were observed in samples collected from pasturelands, such as in Nanshan Pasture, Zhaosu Grassland and Nalat Grasslands, and grass cluster, where a relatively high annual precipitation (>400 mm) and high vegetation coverage were recorded (Fig. 2).

The measured concentrations of $^{137}$Cs vary from (0.92±0.16) Bq kg$^{-1}$ to (31.4±1.7)
Bq kg\(^{-1}\) (Table S1) except samples that were not analyzed due to insufficient amount of samples available for accurate \(\gamma\) measurement, which are similar to the reported concentrations of 4.69-30.1 Bq kg\(^{-1}\) (decay corrected to Sep. 2017) in surface soil samples from adjacent area of 41-42 °N, 86-93 °E in central Xinjiang (Xu et al., 1987). The spatial distribution of \(^{137}\)Cs and \(^{239,240}\)Pu concentrations are similar in this region, both following a decreasing order of pastureland and grass cluster > Gobi and steppe > roads and cultivated lands. This indicates that environmental parameters, such as precipitation, vegetation coverage, soil types, human activities, etc. could directly influence the regional deposition rate and soil retention of these radionuclides, and finally their concentrations and total inventories in the soil column. Similar patterns have been reported in other locations such as Altay region (one of the most affected areas by the SNTS) (Sukhorukov et al., 1999), northeast China (Xu et al., 2013), and Beibu Gulf, China (Xu et al., 2015).

Fig. 2 Concentrations of \(^{239,240}\)Pu in surface soil from 27 sampling sites (near roads and cultivated land, Gobi and desert steppe, Pastureland and grass cluster) in Northern Xinjiang
The $^{240}\text{Pu}/^{239}\text{Pu}$ atomic ratios in the surface soil samples varied from 0.118 to 0.209 (Fig. 3). Significantly lower $^{240}\text{Pu}/^{239}\text{Pu}$ atomic ratios of 0.118-0.133 compared to that of global fallout (0.180±0.014) (Kelley et al., 1999) were obtained in 5 samples in the northwest region of the studied area, indicating a possibly regional deposition of other sources except the global fallout. At two sites in the same region, $^{240}\text{Pu}/^{239}\text{Pu}$ atomic ratios of 0.161±0.067 and 0.166±0.008 were measured, which are similar to the global fallout (0.180±0.014). This might be attributed to the inhomogeneous deposition in the region due to the regional rainfall and topography. Such inhomogeneous distribution pattern of plutonium and cesium have also been reported in surrounding areas of the SNTS (Yamamoto et al., 2004).

Slightly lower $^{240}\text{Pu}/^{239}\text{Pu}$ atomic ratios of (0.152±0.055), (0.153±0.014), and (0.156±0.013) compared to the global fallout were observed in three samples (S101, 103 and 105) from the east part of the studied area, and possible regional deposition from other sources might have a contribution in these sites. Besides the significantly lower values in the northwest part, the $^{240}\text{Pu}/^{239}\text{Pu}$ atomic ratios in the rest samples (range: 0.152-0.209, average: 0.172±0.012) just fall into the range of global fallout level (0.180±0.014), indicating the global fallout is the dominant source of plutonium in these sites.
Fig. 3 Spatial distribution of $^{240}$Pu/$^{239}$Pu atomic ratios in the surface soil from Northern Xinjiang

The measured $^{239,240}$Pu/$^{137}$Cs activity ratios in the surface soils are 0.039-0.215, with two significantly higher values of (0.101±0.003) and (0.215±0.001) in samples from the northwest of the investigated area (S116 and 118), and slightly higher values of (0.065±0.002) in the middle (S112), and (0.081±0.012) in the east region (S106) (Fig. 4). For the remaining sites in Northern Xinjiang, the $^{239,240}$Pu/$^{137}$Cs activity ratios (0.038-0.050) are comparable to the reported value ranges of 0.030-0.048 in soils from the north part of China (37-40 °N) and 0.035-0.051 in the sites of similar latitude in the USA (40 °N), which are mainly affected by the global fallout (Sha et al., 1991; Hodge et al., 1996). The two distinctly higher $^{239,240}$Pu/$^{137}$Cs activity ratios observed in the northwest imply a regional deposition of sources other than global fallout, which also agrees with the observation of $^{240}$Pu/$^{239}$Pu atomic ratios, confirming the deposition of radioactive pollutant other than global fallout in this part. The other two slightly higher
$^{239,240}\text{Pu}/^{137}\text{Cs}$ activity ratios (0.065-0.081) were observed in the middle and east part of the investigated area (S112 and S106), while the $^{240}\text{Pu}/^{239}\text{Pu}$ atomic ratio in these two site do not show lower values compared to the global fallout. The slightly different deposition and migration of plutonium and $^{137}\text{Cs}$ in the environment induced by their different chemical properties, speciation in the soil and environmental behaviors (Qiao et al., 2012) might lead to the slightly different activity ratios of $^{239,240}\text{Pu}/^{137}\text{Cs}$ in these two sites. A more systematic investigation on this issue is needed to clarify it.

![Spatial distribution of $^{239,240}\text{Pu}/^{137}\text{Cs}$ activity ratios in surface soil in Northern Xinjiang](image)

3.2 Depth distribution of $^{239,240}\text{Pu}$ and $^{137}\text{Cs}$ in soil profiles

In the core XJ-01, the maximum $^{239,240}\text{Pu}$ concentration of 0.079 Bq kg$^{-1}$ was measured in the top 5 cm. This value is much lower compared to that measured in other soil profiles in this work (0.160-1.87 Bq kg$^{-1}$), and decreased to even lower values in
deep layers of this soil core. As the soil core XJ-01 was collected near the Barkol Lake, the low Pu concentrations in this core is probably related to direct water erosion of plutonium associated soil particles by periodic water flow intrusions as that reported in Beibu Gulf (Xu et al., 2015). Relatively homogeneous vertical distribution of $^{239,240}$Pu concentrations were observed in the cores of XJ-02 (near a main road) and XJ-07 (in the suburbs of Mori County) with average values of 0.12 Bq kg$^{-1}$ and 0.16 Bq kg$^{-1}$, respectively, indicating significantly vertical mixing of soil in these two cores (Fig. 5).

These three cores are therefore excluded from the further discussion.

![Fig. 5 Vertical distributions of $^{239,240}$Pu concentrations and $^{240}$Pu/$^{239}$Pu atomic ratios in 7 soil cores of XJ-01-07](image-url)
The $^{239,240}$Pu concentrations in the 4 soil cores of XJ-03, 04, 05 and 06 varied from 0.006 to 1.87 Bq kg$^{-1}$, with maximum values in the surface layer and then exponentially decreased along depth (Fig. 5). This trend is similar to undisturbed soil cores observed in Korea (Lee et al., 1996; Meusburger et al., 2016), northeast part of China (Xu et al., 2013), and Australia (Lal et al., 2013), indicating less disturbances in these four soil cores. The $^{239,240}$Pu concentration ranges of the four cores are different, with 0.016-0.430 Bq kg$^{-1}$ in the core XJ-03, 0.010-0.527 Bq kg$^{-1}$ in the core XJ-04, 0.119-1.87 Bq kg$^{-1}$ in the core XJ-05 and 0.009-1.57 Bq kg$^{-1}$ in the core XJ-06. The different concentration ranges of $^{239,240}$Pu in these cores might be attributed to the different sampling sites and relative environment factors. The higher LOI levels in XJ-05 (9.30%) and 06 (5.58%) help reserved plutonium compared to that in XJ-03 (2.45%) and 04 (2.85%) (Zhang and Hou, 2019a). The relatively deep downward migration of Pu was observed in XJ-05 (20 cm) and XJ-03 (15 cm) compared to XJ-04 and 06 (5 cm) (Fig. 5), which matches well with the higher average moisture content in XJ-03 (8.74%) and 05 (10.70%) compared to that in XJ-04 (3.71%) and 06 (1.68%). The high moisture content might speed up the decomposition of organic matters and generate low-molecular weight metabolic products, such as citric acid (Kerner and Edelkraut, 1995; Choppin, 1992), which was observed to form soluble Pu-citrate complexes and facilitate downwards migration of Pu (Fancis et al., 2015). In these four soil cores, most of $^{239,240}$Pu was preserved in the top layers (Fig. 5), only small fraction was migrated to the deep layer (>10 cm). The $^{239,240}$Pu concentrations in deeper layer of the soil core are very low (<0.02 Bq kg$^{-1}$), and consequentially high analytical uncertainties for both
\[ ^{239,240}\text{Pu} \] concentrations (especially the \[^{240}\text{Pu} \] concentrations) and \[^{240}\text{Pu}/^{239}\text{Pu} \] atomic ratios in the deeper layer were obtained, therefore are not suitable for further discussion. The \[^{240}\text{Pu}/^{239}\text{Pu} \] atomic ratios in the upper layers of these 4 soil cores are 0.102-0.156 for XJ-03 (0-10 cm), 0.162-0.209, 0.168-0.178, and 0.173-0.209 for XJ-04 (0-5 cm), XJ-05 (0-20 cm), and XJ-06 (0-5 cm) core respectively. In the core XJ-03, a relative higher ratio of 0.156±0.048 was observed in the surface layer (0-5 cm), while a ratio of only 0.102±0.021 was measured in the 5-10 cm layer, clearly indicating the presence of sources other than the global fallout. In core XJ-05 and upper layer of the cores XJ-04 and XJ-06, the \[^{240}\text{Pu}/^{239}\text{Pu} \] atomic ratios were similar to the global fallout level (Fig. 5). In consideration of the northwest locations of XJ-03, the lower ratio in the subsurface layer might indicate a different source of plutonium except the global fallout. The other three cores located in the south part of the studied area might mainly receive the global fallout derived plutonium. This is similar to the observation in the spatial distribution of \[^{240}\text{Pu}/^{239}\text{Pu} \] atomic ratios in surface soils. Low \[^{240}\text{Pu}/^{239}\text{Pu} \] atomic ratios of less than 0.10 were observed at 5.5 cm and 7.5 cm layers in soil core XJ-04, which is significantly lower than the value of global fallout, the contribution from other source such as the SNTS cannot be excluded. Low \[^{240}\text{Pu}/^{239}\text{Pu} \] ratios of 0.10-0.12 at 13-17 cm depth in the soil core XJ-06 were also observed. However, the \[^{239,240}\text{Pu} \] concentrations decreased to less than 0.014 Bq kg\(^{-1}\) below 5 cm in XJ-04, and to less than 0.024 Bq kg\(^{-1}\) below 9 cm in the core XJ-06. Accordingly, the analytical uncertainties for \[^{240}\text{Pu} \] in these deep layer soil samples were high, causing large uncertainties of \[^{240}\text{Pu}/^{239}\text{Pu} \] atomic ratios. Therefore, it is difficult to attribute the
relatively lower $^{240}$Pu/$^{239}$Pu ratios in some deep layer of these two cores to the
collection from the other sources except the global fallout.

Small variations of $^{240}$Pu/$^{239}$Pu atomic ratios were observed along the depth in the
cores XJ-02 (0.167-0.186) and XJ-07 (0.158-0.167), which are close to the mean global
fallout value and similar to those observed in the nearby surface soil, indicating the
global fallout deposition is the dominant source of plutonium in the southern part of the
investigated area. Due to the low concentrations of $^{239,240}$Pu along the core XJ-01, the
obtained $^{240}$Pu/$^{239}$Pu atomic ratios are not reliable, which is therefore excluded from
discussion.

The concentrations of $^{137}$Cs in some core samples from XJ-01, 03, 04 and 06 were
lower than the detection limit (1 Bq kg$^{-1}$). Besides the relatively short half-life of $^{137}$Cs
($T_{1/2} = 30.2$ yr) and the radioactive deposition in this region mainly happened before
1980 (40 years ago), the disturbance of sampling site near the Barkol Lake (XJ-01) and
soil erosion at site XJ-03 near a main road (details discussion in 4.1) are the main
reasons causing low concentrations of $^{137}$Cs in these samples. The concentrations of
$^{137}$Cs in the cores of XJ-04 and 06 decline rapidly from the surface to deep layers and
only data in the 0-4 cm and 0-2 cm were determined in these two cores, respectively.
Finally, only data from the cores XJ-02, 05 and 07 were used to draw the depth
distribution of $^{137}$Cs concentrations and $^{239,240}$Pu/$^{137}$Cs activity ratios (Figure S1).

The $^{239,240}$Pu inventories in the soil cores estimated by integrating $^{239,240}$Pu content
in all layers from each core are 88.4±10.1 Bq m$^{-2}$ for XJ-3, 86.1±6.9 Bq m$^{-2}$ for XJ-04,
145±15 Bq m$^{-2}$ for XJ-05 and 112±20 Bq m$^{-2}$ for XJ-06 core (Fig. 5). The varied
inventories of plutonium at different sites are mainly attributed to the tomography of
the sampling site, local climate, properties of soil such as LOI and moisture content,
these factors influence the reservation of plutonium in the soil, and also the amount of
deposition. The estimated inventories of $^{239,240}\text{Pu}$ in these soil cores are comparable to
the reported values in Beijing (39.9°N: 107 Bq m$^{-2}$), Dalian (39.0°N: 84.5-90.0 Bq m$^{-2}$),
Chengde (40.9°N: 138 Bq m$^{-2}$), and Qingyang (35.7°N: 110 Bq m$^{-2}$) (Dong, 2010b;
Xu et al., 2015; Ni et al., 2018; Zhang et al., 2019b) in the north of China, but much
lower than the values of 109-546 Bq m$^{-2}$ reported in Guazhou and Dunhuang city that
located in the downwind area of Lop Nor nuclear weapons testing site (Bu et al., 2015),
and 120-200 Bq m$^{-2}$ in the downwind area of the SNTS (Yamamoto et al., 2004) and
40900 Bq m$^{-2}$ in the supposed center-axis of one radioactive plume from the SNTS
(Yamamoto et al., 2008).

4. Discussion

4.1 Level of radioactive pollution in the studied area

The measured concentrations of $^{239,240}\text{Pu}$ (0.77-1.20 Bq kg$^{-1}$) and $^{137}\text{Cs}$ (0.92-31.4
Bq kg$^{-1}$) in the surface soil collected in Northern Xinjiang fall into the reported levels
in the background areas (no direct contamination from nuclear activities and accidents,
$^{239,240}\text{Pu}$: 0.37-3.7 Bq kg$^{-1}$, $^{137}\text{Cs}$: 0.70-41.26 Bq kg$^{-1}$, decay corrected to Sep. 2017) (Xu
et al., 2013; Lee et al., 1996; Zheng et al., 2008). The estimated $^{239,240}\text{Pu}$ inventories in
the soil cores XJ-04 (86.1±6.9 Bq m$^{-2}$), XJ-05 (145.1±15.1 Bq m$^{-2}$) and XJ-06
(112.1±20.4 Bq m\(^{-2}\)) are also similar to the reported values in the background area (90.0-138.0 Bq m\(^{-2}\)) (ATSDR, 2010; Dong, 2010b; Xu et al., 2015; Ni et al., 2018; Zhang et al., 2019b). These results indicate that the level of radioactive pollution in this area falls into the global background level.

However, significantly lower \(^{240}\text{Pu}/^{239}\text{Pu}\) atomic ratios were measured in 5 surface soil (0.118-0.133) in the northwest part of Northern Xinjiang, which are different from that of global fallout ratios (0.180±0.014). This implies that besides the global fallout other sources also had a remarkable contribution to plutonium in this region. While the total inventory of \(^{239,240}\text{Pu}\) in the soil core XJ-03 (88.4±10.1 Bq m\(^{-2}\)) is lower than that in the soil cores of XJ-05 and XJ-06 (112-145 Bq m\(^{-2}\)) and in Beijing (39.9 °N: 107 Bq m\(^{-2}\), Chengde (40.9 °N: 138 Bq m\(^{-2}\)) and Qingyang (35.7 °N: 110 Bq m\(^{-2}\)) (Dong, 2010b; Ni et al., 2018; Zhang et al., 2019b). This seems contradictory with the conclusion that both global fallout and other regional sources contributed to the total radioactivity in this region.

The exponentially declining \(^{239,240}\text{Pu}\) concentrations with depth in the soil core XJ-03 and comparable low concentration of \(^{239,240}\text{Pu}\) (0.017 Bq kg\(^{-1}\)) in the deep layer (15-25 cm) compared with other soil cores (XJ-05 and XJ-06) indicate that all plutonium in the soil column was integrated in the total inventory (Fig. 5). However, a relative low concentration of \(^{239,240}\text{Pu}\) (0.43 Bq kg\(^{-1}\)) in the top layer (0-5 cm) was observed, which is about 3 times lower than that measured in the top layer (0-5 cm) from XJ-05 (1.87 Bq kg\(^{-1}\)) and XJ-06 (1.57 Bq kg\(^{-1}\) in the layer of 0-2 cm). This implies that the erosion of surface soil at the site of core XJ-03 might occur, causing loss of the plutonium
deposited on the top of soil profile.

After deposition, plutonium bound to fine particles in the surface soil might be redistributed with the soil (Alewell et al., 2017). The vegetation coverage at the site of soil core XJ-03 is only about 30% compared to 40-100% in the other sites, making the soil grains easy to be removed. In addition, the strong wind in this region will also facilitate the erosion of previously plutonium contaminated soil, causing a relative low level of inventory of radioactivity in the soil core (Pan and Tian, 2014). A similar soil redistribution process has been reported several times at sites near the location of XJ-03 core (Liu et al., 2010; Dawut and Abudushalik, 2010). The LOI value (roughly represents organic content) in XJ-03 (2.45%) is quite low compared to 2.85%-9.30% in the other soil cores (Table 1), which leads to weakly bound plutonium on soil (Zhang and Hou, 2019a). This might be another factor that facilities the loss of plutonium from soil grains, consequentially leading to a low radioactivity inventory at this site.

4.2 Sources of radioactive pollution in Northern Xinjiang

The significantly lower $^{240}\text{Pu}/^{239}\text{Pu}$ atomic ratios of 0.118-0.133 compared to that of global fallout (0.180±0.014), and higher $^{239,240}\text{Pu}/^{137}\text{Cs}$ activity ratios of 0.101-0.215 compared to that of global fallout (0.030-0.051 in the studied latitude belt) (Kelley et al., 1999; Sha et al., 1991; Hodge et al., 1996) were observed in the northwest part of Northern Xinjiang. In the samples from other sites of the investigated area, the measured $^{240}\text{Pu}/^{239}\text{Pu}$ atomic ratios and $^{239,240}\text{Pu}/^{137}\text{Cs}$ activity ratios are comparable to the value of the global fallout. This data clearly show that besides global fallout of
nuclear weapons tests in 1945-1980, other sources with lower $^{240}\text{Pu}/^{239}\text{Pu}$ atomic ratios and higher $^{239,240}\text{Pu}/^{137}\text{Cs}$ activity ratios contributed to the radioactive pollution in the northwest part of Northern Xinjiang. The possible sources in this region include: (1) close-in fallout from nearby atmospheric NWTs; (2) release from NFRPs; (3) release from nuclear accidents; (4) release from the operation of nuclear power plants.

As a particle active radionuclide, plutonium was seldom released from NFRPs to the atmosphere (ATSDR 2010). In addition, the NFRPs at Sellafield in UK and La Hague in France are far away from the investigated region, and the small Chinese NFRPs located in Jiuquan is about 1100-1400 km southeast of the investigated region with less north or northwest wind in this region. It is therefore unlikely to contribute to the plutonium and $^{137}\text{Cs}$ in Northern Xinjiang.

The nuclear accidents at Chernobyl in 1986 and Fukushima in 2011 have also released small amount of plutonium to the environment with high $^{240}\text{Pu}/^{239}\text{Pu}$ atomic ratios of 0.403-0.412 and 0.323-0.330, respectively (Zheng et al., 2012; UNSCEAR 2000; Men et al., 2018). While, much low $^{240}\text{Pu}/^{239}\text{Pu}$ atomic ratio was measured in the soil in Northern Xinjiang. Meanwhile, particle associated plutonium was mostly deposited in surrounding areas of the Chernobyl, although some volatile radionuclides ($^{137}\text{Cs}$, $^{131}\text{I}$, and $^{129}\text{I}$) were detected in North China with low levels (Paatero et al., 2010; Xu et al., 1989; Fan et al., 2016). No corresponding Chernobyl derived plutonium were observed in China and Japan (Wu et al., 2010; Hirose et al., 2003). It was estimated that 4 orders of magnitude less amount of $^{239,240}\text{Pu}$ ($1.0 \times 10^9$ Bq) was released from Fukushima accident comparted to that from Chernobyl accident ($7.2 \times 10^{13}$ Bq) (Zheng
et al., 2012). The Fukushima derived plutonium was only observed in some samples in the 30 km exclusive zone of Fukushima Daiichi Nuclear Power Plant (Zheng et al., 2012). Therefore, the contribution from nuclear accidents to plutonium in Northern Xinjiang is negligible. The anthropogenic uranium isotopes (e.g. $^{236}$U, $^{233}$U) were demonstrated as a useful tracer, especially for tracing reactor sourced radioactive pollution (with very low $^{233}$U/$^{236}$U ratio in reactor fuel originated sources compared to nuclear weapons testing) (Hain et al., 2020). In combination with plutonium isotopes, it might present a more reliable information. A further investigation of uranium isotopes in soils of this region is planned.

There are no nuclear power plants and other nuclear facilities in this region, and even in the east coastal areas of China where most Chinese nuclear power plants are located, no relative plutonium was detected (Zhang and Hou, 2019a). This source can be also excluded.

The Chinese nuclear weapons testing sites at Lop Nor is located in the southeast of the investigated region in Northern Xinjiang with a distance of 400-800 km. In 1964-1980, a certain number of plutonium weapons were detonated in atmosphere at this site (UNSCEAR 2000). The $^{240}$Pu/$^{239}$Pu atomic ratio in the soil samples from the south part of Northern Xinjiang (the closest area to Lop Nor) were measured to be 0.171±0.016, which is close to the value of global fallout. The prevailing winds in Lop Nor are featured with strong eastern Northeast and western Southwest direction in the near surface, and west wind in the middle-high altitude (Zhao et al., 2019), which could not transport the released radioactive substances to Northern Xinjiang. Meanwhile,
Tianshan Mountain with an average altitude of 5000 m, located between Lop Nor and Northern Xinjiang, acts as a natural barrier to prevent the dispersion of the radioactive substances from Lop Nor to the investigated area. The total α activities of plutonium (mainly including $^{238}$Pu, $^{239}$Pu and $^{240}$Pu) measured in milk ((1.84±0.49)$\times 10^{-3}$ Bq kg$^{-1}$), vegetation ((1.86-8.40)$\times 10^{-3}$ Bq kg$^{-1}$) and surface soil samples ((0.242-0.448) Bg kg$^{-1}$) from the south part of studied area (43-44 °N, 85-87 °E) (Zhang et al., 1988a; Xu et al., 1987; Zhang et al., 1988b) during the test period of Lop Nor are comparable to the level in the background area. Comparable $^{240}$Pu/$^{239}$Pu atomic ratios with the reported global fallout values were measured during 1960s-1970s in an ice core collected from the north slope of Tianshan Mountains (Wang et al., 2017), indicating no significant contribution from atmospheric nuclear weapons tests at Lop Nor NWTS to Northern Xinjiang.

The Semipalatinsk nuclear testing site (SNTS) is located only about 700 km northwest of the investigated area in Northern Xinjiang, where 86 air and 30 surface nuclear weapons tests were conducted during the period of 1949-1962 with a total yield of 5.89 Mt (UNSCEAR 2000). The typical $^{240}$Pu/$^{239}$Pu atomic ratios in the close-in fallout at the SNTS were reported to be 0.025-0.072 (Yamamoto et al., 2002; Sakaguchi et al., 2006), which is significant lower than the global fallout featured $^{240}$Pu/$^{239}$Pu atomic ratios (0.180±0.014). The typical $^{239,240}$Pu/$^{137}$Cs activity ratios in the close-in fallout from the SNTS were observed to be 1.25-1.87 (decay corrected to 2017) (Yamamoto et al., 2002; Sakaguchi et al., 2006), which is significant higher than the global fallout featured $^{239,240}$Pu/$^{137}$Cs activity ratios (0.030-0.051). The reduced $^{240}$Pu/$^{239}$Pu ratios and increased $^{239,240}$Pu/$^{137}$Cs activity ratios in the northwest of
Northern Xinjiang are therefore attributed to the contribution from the regional fallout of the SNTS. Both the direct transport of radioactive plumes released during the tests and re-suspended radioactive materials from the close-in area of detonation sites after the tests could impact the northwest part of the studied area. Since the low burst height and relatively small explosion yields of most weapons tests, the radioactive substances released from the SNTS were mainly deposited in the local area and downwind regional areas (UNSCEAR 2000). The major radioactive plumes were traced to move to the east, northeast, south and southeast directions from the testing site, of them the ones conducted on 12\textsuperscript{th} Aug. 1953 (40 kt fission), 30\textsuperscript{th} Oct. 1954 (55 m, 10 kt) and 24\textsuperscript{th} Aug 1956 (93 m, 27 kt) directly moved to the southeast (Sukhorukov et al., 1999; Yamamoto et al., 2004). The investigated area is located in the downwind direction of the SNST, therefore, directly exposed to the radioactive plume of these weapons tests. The radioactive pollution including plutonium in Northern Xinjiang should be mainly originated from the releases of those nuclear weapons tests at SNTS besides the global fallout.

4.3 Transport pathways and contributions from the SNTS

The radioactive substances released by the nuclear weapons testing at the SNTS was transported to Northern Xinjiang through atmospheric dispersion, the suitable topographic and weather condition might cause its deposition in the northwest part of this region.

The mid-latitude Westerlies dominate the major tropospheric circulation in the
Eurasian center including the SNTS and Northern Xinjiang (Bothe et al., 2012). With the Tarbagatai Mountains in the south (lie west-east) and Altai Mountains in the north (lie northwest to southeast), a valley of trumpet-shaped topography is formed between them (Fig. 1). The Irtysh River originates from the southwest slope of the Altai Mountains, and runs northwest along this valley, through the northwest part of the investigated region and the SNTS, finally flows to the Arctic Ocean. A much lower altitude of 500 m along the Irtysh River valley compared to that over 2000 m in the sideward mountains introduce the Westerlies into this area (Tian et al., 2007). Additionally, due to the perennial Siberian cold high-pressure air in the northwest, frequent west and northwest winds (>61%) occur in the northwest of Northern Xinjiang (Habahe County, Jimunai County, and Burqin County) (Pan and Tian, 2014). The forward trajectories analysis of air masses by the HYSPLIT model shows that the air masses in troposphere after some nuclear weapons tests at SNTS arrived the northwest part of the investigated area in Northern Xinjiang (Fig. S2), such as the plumes at 2000-4000 m height after the test on 12th Aug. 1953, the plumes at 3000 m and 4000 m height after the test on 30th Oct. 1954, and the plumes at 2000-4000 m height after the test on 24th Aug. 1956. Therefore, the southeastwards dispersion of radioactive plumes released from the SNTS directly moved to the northwest part of the investigated region in Northern Xinjiang, which might be the major transport pathway. The measured $^{240}\text{Pu}/^{239}\text{Pu}$ atomic ratios in the soil samples from the south of this area are close to the global fallout level, indicating that the radioactive plume from the SNTS might not transport further southwards.
A relatively high annual precipitation rate of 190-700 mm yr\(^{-1}\) was reported in the northwest part of Northern Xinjiang (Li et al., 2015), which promotes the deposition of the radioactive substance carried by the air masses. This might also explain the high deposition of the SNTS derived radioactive substance in this area but not further downwind area. In addition, the three important weapons tests with dominant southeastwards plumes were all conducted in late summer to autumn (August and October), i.e. the high precipitation season according to the metrological data records (Chen et al., 2015), which intensively increase the washout rate of particles associated plutonium from the atmosphere to the land in this area. The analysis of plutonium isotopes in an ice core collected from the Belukha glacier in Altai Mountains (<200 km to the northwest of studied area) showed low \(^{240}\text{Pu}/^{239}\text{Pu} \) atomic ratios of 0.08 and 0.15 in the segments of 1949-1952 and 1952-1955 respectively, which further confirmed the radioactive source from the SNTS in this area (Olivier et al., 2004).

Based on the above discussion, the radioactive pollution in Northern Xinjiang, especially the northwest part, is originated from both regional fallout of the SNTS and the global fallout, and the records of plutonium in the soil are a mixture of the two sources. The contribution of each source can be estimated based on the characteristic atomic ratios of \(^{240}\text{Pu}/^{239}\text{Pu} \) from the global fallout and close-in fallout of the SNTS. The empirical binary mixing model was used (Hardy 1976; Krey and Hardy, 1970), and shown below:

\[
Y = \frac{(R_G - R_S)}{(R_S - R_W)} \cdot \frac{(1 + 3.73R_W)/(1 + 3.73R_G)}
\]

Where \(R_S\)-atomic ratios of \(^{240}\text{Pu}/^{239}\text{Pu} \) in the sample, \(R_G\)-atomic ratios of \(^{240}\text{Pu}/^{239}\text{Pu} \)
of global fallout, RW-atomic ratios of $^{240}$Pu/$^{239}$Pu in close-in fallout from the SNTS,

Y-contribution ratio of regional fallout $^{239,240}$Pu to global fallout $^{239,240}$Pu.

Of the above parameters, $R_S$ is obtained through the sample measurements. Here, the distinctly lower ratios of 0.118-0.133 measured in the northwest part of the studied area are used. $R_G$ is well confirmed to be 0.180. Since various types of nuclear weapons tests with different magnitudes were conducted at the SNTS, the atomic ratios of $^{240}$Pu/$^{239}$Pu were reported to vary within 0.025-0.072 (Yamamoto et al., 2002).

According to the measurement of $^{240}$Pu/$^{239}$Pu atomic ratios in the territory of the SNTS, about 77-95% of the ratios were in the range of 0.03-0.05 (Yamamoto et al., 2002). Therefore, the lowest ratio of 0.025 was widely used as the typical $^{240}$Pu/$^{239}$Pu atomic ratio from the SNTS (Yamamoto et al., 2002; Yamamoto et al., 1999). With these data, the regional contribution from SNTS of 28-43% compared to the global fallout input are estimated in the northwest of Northern Xinjiang.

5. Conclusion

Based on the determined $^{239}$Pu, $^{240}$Pu and $^{137}$Cs concentrations in surface soil and soil profiles collected in Northern Xinjiang, it can be concluded that (1) the $^{239,240}$Pu concentrations vary within 0.06-1.20 Bq kg$^{-1}$, and $^{137}$Cs in <1-31.4 Bq kg$^{-1}$, the $^{239,240}$Pu inventories in the soil cores are 86.1-145 Bq m$^{-2}$, all falling into the range of background level, indicating no serious radioactive contamination in Northern Xinjiang; (2) distinctly lower $^{240}$Pu/$^{239}$Pu atomic ratios of 0.118-0.133 and higher $^{239,240}$Pu/$^{137}$Cs activity ratios of 0.065-0.215 compared to the global fallout values were observed in
the northwest part of Northern Xinjiang, indicating other sources of radioactive
deposition in this area besides the global fallout; (3) regional fallout of nuclear weapons
test in SNWT has a significant contribution of the anthropogenic radioactivity in
northwest part of Northern Xinjiang; 4) the Westerlies carried radioactive substance
released from the SNTS enters into the northwest part of Northern Xinjiang through a
trumpet-shaped topography along the Irtysh River Valley. The contribution from the
SNTS was estimated based on the measured $^{240}\text{Pu}/^{239}\text{Pu}$ atomic ratios to be 28-43% of
the global fallout amount.

Acknowledgements

This work was supported by the Ministry of Science and Technology of China [No.
2015FY110800], the Natural Science Foundation of China [No. 11605207, 11605206,
11875261, 91643206 and No. 41991252], Chinese Academy of Sciences (No.
132B61KYSB20180003 and No. XDB40020104), and the State Key Laboratory of
Loess and Quaternary Geology. X. Zhao thanks to Zhao Huang, Weichao Zhang and
Yanyun Wang from the Institute of Earth Environment for the field sampling work and
graphing instructions, and Liuchao Zhu from the Technical University of Denmark for
his help in the measurement of plutonium.

Reference

Alewell, C., Pitois, A., Meusburger, K., Ketterer, M., Mabit, L., 2017. $^{239+240}\text{Pu}$ from “contaminant”to


Guli, J., Liang, S., Yi, Q., Liu, J., 2015. Vegetation dynamics and responses to recent climate change
in Xinjiang using leaf area index as an indicator. Ecol. Indic. 58, 64-76.


Sha, L., Yamamoto, M., Komura, K., Ueno, K., 1991. $^{239,240}$Pu, $^{241}$Am and $^{137}$Cs in soils from several areas in China. J. Radioanal. Nucl. Ch. 155, 45-53.


Xu, H., Wei, J., Yan, Z., Zou, W., 1989. 131I level in milk, goat milk, breast milk and pasture samples after the Chernobyl and the relevant contamination evaluation. CN. J. Public Health 5, 18-21.


Yamamoto, M., Tomita, J., Sakaguchi, A., Imanaka, T., Fukutani, S., Endo, S., Tanaka, K., Hoshi, M., Gusev, B. I., Apsalikov, A. N., 2008. Spatial distribution of soil contamination by $^{137}$Cs and
\(^{239,240}\)Pu in the village of Dolon near the Semipalatinsk nuclear test site: new information on traces of the radioactive plume from the 29 August 1949 nuclear test. Health Phys. 94, 328-337.


