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Particle-size speciation of Pu isotopes in surface soils from Inner Mongolia (China) and its implications for Asian Dust monitoring

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ABSTRACT

To study the applications of Pu isotopes in long-distance dust migration monitoring, Pu isotopes in surface soil of Inner Mongolia have been analyzed using SF-ICP-MS after size fractionation. $^{240}Pu/^{239}Pu$ atom ratios ranged narrowly (0.169–0.200) and indicated global fallout character, while $^{239+240}Pu$ activities increased with decreasing particle size. A spherical model could well simulate $^{239+240}Pu$ activities as a function of particle diameter when soil particle size was less than 600 μm, and the soil particle surface sorption phenomenon of Pu isotopes in natural soil samples was revealed. Furthermore, $^{239+240}Pu$ activity in fine particles (sized less than 53 μm) had good consistency with that in atmospheric depositions of Japan since the 2000s, suggesting new Asian Dust sources (i.e. central Inner Mongolia) other than the well-known Chinese deserts.

1. Introduction

Plutonium isotopes, characterized by high chemical toxicity and long half-lives, have entered the environment mainly due to atmospheric nuclear weapon tests during the period from the 1950s to the early 1960s, failed launches of nuclear-battery-powered satellites and nuclear reactor accidents (Harley, 1980). A total of 543 atmospheric nuclear tests took place worldwide from 1945 to 1980, including smaller scale atmospheric nuclear tests conducted by France and China in the post-Test Ban Treaty era after 1962 (UNSCEAR, 2000). The two well-known nuclear reactor accidents, Chernobyl accident in 1986 and Fukushima Daiichi Nuclear Power Plant accident in 2011, were estimated to release 8.7×10^{13} Bq and 1.1×10^{9}-2.4×10^{9} Bq $^{239+240}Pu$ to the environment, respectively (UNSCEAR, 2008; Zheng et al., 2012). As far as we knew, to date, no Pu isotope information related to nuclear weapon tests of North Korea has been published.

In the past decades, Pu isotopes have gradually deposited on the earth's surface through wet and dry deposition patterns locally, regionally and globally. Pu distributions in land surfaces across many countries have been determined and published, but reports of Pu distributions in China are scarce and those that are available are limited. Pu information for soil samples around the Chinese Nuclear Test Site (CNTS, Lop Nor, see Fig. 1a) was first reported by Zhang et al. (1988) and Lin et al. (1992). After that, Zheng et al. (2009) studied $^{239+240}Pu$ activities and $^{240}Pu/^{239}Pu$ atom ratios in soil samples of northwestern China (Gansu) using ICP-MS, while Dong et al. (2010) studied those activities and ratios in soil samples of central (Hubei) China using AMS. Comprehensive research on Pu isotopes of loess in the downwind areas of the CNTS has been conducted by Bu et al. (2015, 2016) to elucidate local fallout of Chinese nuclear testing. Global fallout Pu in samples were examined by Bu et al. (2014) in soil samples of southwestern China (Chongqing and Guizhou) and by Xu et al. (2013) in soil samples of northeastern China (Liaodong Bay); these authors were studying migration behaviors of Pu isotopes and evaluating soil erosion rates. To the best of our knowledge, no study focusing on Pu isotope information in Inner Mongolia has been conducted.

The deteriorated soil erosion and associated sedimentation arising from human activities have become a serious problem in many developing countries including China. With similar environmental behaviors to those of ^{137}Cs, fallout Pu isotopes have been used as tracers, instead of short-lived radiocesium, in studies about ongoing environmental changes such as land degradation and desertification followed by dust storms. According to the long-term anthropogenic radionuclide deposition monitoring conducted by the Meteorological Research Institute in Japan since 1956, the decline in the anthropogenic radionuclide deposition slowed or almost ceased after 1988, which implied that resuspension of radionuclides deposited on land started to play an important role in current rates of radionuclide deposition

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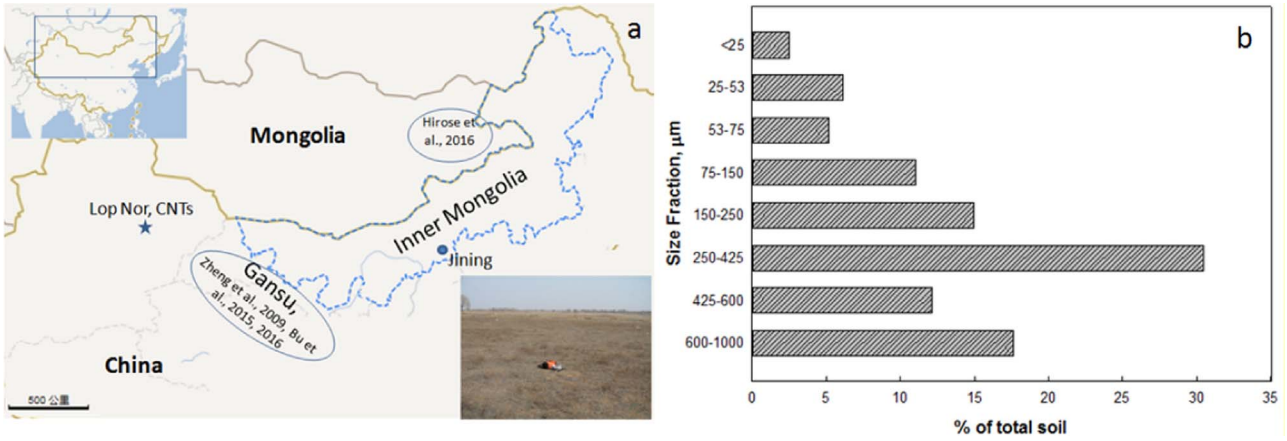

Fig. 1. (a) Map showing the soil sampling site in Jining, central Inner Mongolia, with adjacent sampling regions reported in the literature (Zheng et al., 2009; Bu et al., 2015, 2016; Hirose et al., 2016) and CNTS at Lop Nor. (b) Particle size distribution of the surface soil sample.

(Igarashi et al., 2001; Hirose et al., 2003; Lee et al., 2006). Further studies indicated that long-range transport of suspended soil dust produced in Asian deserts and arid regions affected deposition in Japan, especially in springtime events known as Asian Dust storms (also referred to as Kosa events or yellow dust storms) (Hirose et al., 2007). From the 2000s, monitoring of anthropogenic nuclides in the atmospheric depositions in Japan has suggested possible change of the Asian Dust source regions: shifting from the arid zone to the desert-steppe zone that is undergoing desertification (Igarashi et al., 2009). Therefore, it is important to have a better understanding of Pu isotopes in the surface soil of Inner Mongolia because the North China Plain, including central Inner Mongolia, was also recognized as one potential new source region of the Asian Dust (Igarashi et al., 2009, 2011).

In this work, we have studied activities, isotope ratios, and particle size distributions of Pu isotopes in surface soil samples collected from central Inner Mongolia in order to: (1) establish the Pu database for Inner Mongolia that can be used to make possible source identifications and reasonable comparisons with results of adjacent regions; (2) study the particle size distribution of Pu isotopes in environmental soil samples; and (3) investigate resuspension and long-term transportation of Pu isotopes by Asia Dust and try to clarify the possible shifting of the source region of the Asian Dust.

2. Experimental

The sampling location is shown in Fig. 1a. Surface soil samples (0–5 cm) were collected in April 2009 from undisturbed grassland of Jining, which is located in central Inner Mongolia (40.95°N, 113.21°E). More than 5 samples were collected randomly, with a separation distance of more than 50 m from each other and mixed. Inner Mongolia is a climatic transition zone between the arid and semi-arid northwest inland region and the humid and semi-humid southeast coastal region that is affected by the Asian monsoon. The climate in Inner Mongolia is cold and dry in winter, and warm and wet in summer. The annual precipitation of Jining ranges from 150 to 450 mm with the average value of 393 mm (Ha et al., 2006). (Data from the website of local government: http://www.wulanchabu.gov.cn).

After drying the mixed samples at 110 °C for at least 8 h, size factions were determined by mechanically shaking a sample (about 40 g) for 3 h to pass through sequential sieves with diameters ranged from 25 μm to 1 mm. As shown in Fig. 1b, the size fraction of the largest amount was coarse, 250–425 μm.

The chemical procedures for Pu separation in soil samples were based on our previous work (Zheng et al., 2009). Briefly, a sample of 1–3 g was weighed out and spiked with 1 pg ^{242}Pu as yield monitor. After total digestion with $HNO_3 + HF + HClO_4$, Pu isotopes were co-precipitated with Fe^{3+} ions and hence separated and purified using two-stage anion-exchange chromatography. We used SF-ICP-MS (Element 2, Thermo Finnigan, Bremen, Germany) for the analysis of Pu isotopes. The detailed operational setup and parameters of this analytical system for Pu analysis have been given elsewhere (Zheng et al., 2006).

3. Results and discussion

3.1. Pu isotopes in surface soil of central Inner Mongolia

Our results of $^{239+240}$Pu activities and ^{240}Pu/^{239}Pu atom ratios in surface soil samples collected in central Inner Mongolia are presented in Table 1 and Fig. 2. The $^{239+240}$Pu activity of the surface soil ranged from 0.101 ± 0.012 mBq/g to 0.519 ± 0.022 mBq/g depending on soil particle size fractions, with the mass-weighted activity of 0.231 ± 0.014 mBq/g. As shown in Fig. 2, $^{239+240}$Pu activities gradually decreased with increasing particle size and $^{239+240}$Pu activities in the fine particles are markedly higher than those of coarse particles.

It was difficult to conduct direct comparisons with our results with previous work by other scientists because different sample preparations can affect the results significantly, such as different particle size-fractionated surface soil samples selecting, or acid leaching v.s. total digestion method. Thus, we performed careful comparisons with previously published $^{239+240}$Pu activities of surface soils in adjacent regions. Using exactly the same preparation procedures but simply separating surface soil into a coarse (> 150 μm) and a fine (< 150 μm) fraction, Zheng et al. (2009) reported roughly comparable but half as large $^{239+240}$Pu activities in the samples from Gansu in northwestern China (0.005–0.157 mBq/g for < 150 μm) as ours (0.326 ± 0.015 mBq/g for < 150 μm), which was probably due to a faster migration rate to deeper soil layers as well as wind erosion, considering the similar low annual precipitation (235–328 mm) and uniquely low organic content in loess of Gansu. The average $^{239+240}$Pu activity

Table 1
$^{239+240}$Pu activities and ^{240}Pu/^{239}Pu atom ratios in surface soil of Inner Mongolia, China.

Size fraction (μm)	$^{239+240}$Pu activity (mBq/g)	^{240}Pu/^{239}Pu atom ratio
600–1000	0.101 ± 0.012	0.199 ± 0.051
425–500	0.119 ± 0.011	0.200 ± 0.029
250–425	0.256 ± 0.017	0.169 ± 0.023
150–250	0.265 ± 0.009	0.190 ± 0.013
75–150	0.252 ± 0.014	0.174 ± 0.016
53–75	0.340 ± 0.015	0.189 ± 0.015
25–53	0.371 ± 0.015	0.194 ± 0.018
< 25	0.519 ± 0.022	0.181 ± 0.020
Average	0.231 ± 0.014 (mass-weighted)	0.182 ± 0.021 (activity-weighted)

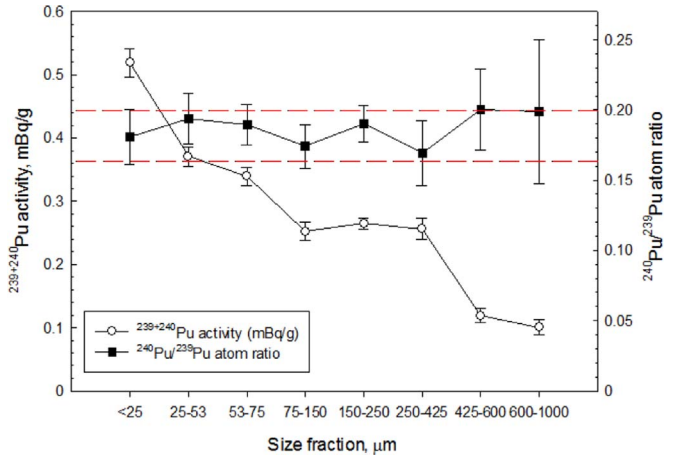

Fig. 2. Distributions of $^{239+240}$Pu activities and ^{240}Pu/^{239}Pu atom ratios in particle size-fractionated surface soil samples from central Inner Mongolia, China. The dashed lines indicate the global fallout range (0.180 ± 0.014) in the northern hemisphere (Kelley et al., 1999) (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)

obtained in this study was also similar to that of downwind areas of the CNTS (0.365 mBq/g), also in northwestern China, although large variation of $^{239+240}$Pu activities depending on sample locations was observed (0.023–1.988 mBq/g) (Bu et al., 2015, 2016). Most recently, Hirose et al. (2016) investigated Pu isotopes in Mongolian grassland surface soils (only < 53 μm fraction) and obtained an activity range of 0.42 ± 0.03 to 3.53 ± 0.09 mBq/g, systematically higher than our observations of the same fraction (0.414 ± 0.017 mBq/g). Results in this study were also comparable with that of northeastern China (0.023–0.938 mBq/g) and our previous work of central China (0.358–0.380 mBq/g), although different preparations and measurement methods were performed (Dong et al., 2010; Xu et al., 2013).

Unlike the significant difference of $^{239+240}$Pu activities seen between the coarse particles and the fine ones, ^{240}Pu/^{239}Pu atom ratios did not show any obvious variation between them: they ranged narrowly from 0.169 ± 0.023 to 0.200 ± 0.029 with the activity-weighted ratio of 0.182 ± 0.021. As shown in Fig. 2, higher uncertainties for ^{240}Pu/^{239}Pu atom ratios of the coarse particles (particle size > 240 μm) were observed. This is due to the decrease of $^{239+240}$Pu activities with increasing of particle size, resulting in the decrease of Pu isotopes' counts during the measurements. The observed ^{240}Pu/^{239}Pu atom ratios in the present study were consistent with the global fallout value of 0.180 ± 0.014 in the northern hemisphere (Kelley et al., 1999). Similar global fallout results have been found in soil samples of adjacent areas: 0.169–0.192 with an average of 0.182 ± 0.008 (Gansu) (Zheng et al., 2009), 0.144–0.245 with an average of 0.192 ± 0.020 (Liaodong Bay) (Xu et al., 2013) and 0.172–0.220 (Hubei) (Dong et al., 2010). Bu et al. (2015, 2016) identified lower ^{240}Pu/^{239}Pu atom ratios than the global fallout ratio in downwind areas of the CNTS, indicating a mixed fingerprint of Pu from the CNTS. However, the great distance (~2000 km) between our present sampling site and the CNTS might indicate that the direct contribution of the close-in fallout from the CNTS to Pu in central Inner Mongolia should be negligible. Based on these facts, we can conclude that the dominate source of Pu isotopes in central as well as eastern Inner Mongolia is likely to be the global fallout of atmospheric nuclear tests, although that of western Inner Mongolia still needs further verifications.

3.2. Distribution of Pu isotopes as a function of soil particle size

As shown in Table 1, the $^{239+240}$Pu activities in the finest particles (< 25 μm, 0.519 ± 0.022 mBq/g) were nearly 5 times higher than that in the coarse particles (600–1000 μm, 0.101 ± 0.012 mBq/g). For the other fractions, $^{239+240}$Pu activities clearly increased with decreasing particle radius. Lee et al. (2004) indicated that Pu and other actinides tend to partition to certain soil particles rather than sorb homogeneously to all particles, which reflects that their association is a surface sorption phenomenon. Further, they indicated the increasing activities of $^{239+240}$Pu in smaller size fractions probably are due to the greater surface area provided for sorption. Baeza et al. (1995) have established a simple but practical spherical model to link activity with particle sizes. The total activity (in Bq), A_{total}, of a particle can be expressed as:

$$A_{total} = A_{surface} + A_{volume} = k_1 S + k_2 V \quad (1)$$

where k_1 is the surface activity (Bq/m^2) and k_2 is the activity per unit volume (Bq/m^3), S and V are surface area and volume of the particle, respectively. Assuming the particle to be spherical, mass is M and density is d, Eq. (2) can express the massic activity of isotopes (in Bq/kg):

$$A_{massic} = \frac{A_{total}}{M} = (k_1 S + k_2 V)/dV \cong 3k_1/d \times R + k_2/d = P_1/R + P_2 \quad (2)$$

where P_1 is $3k_1/d$ and P_2 is k_2/d, and are parameters. Eq. (2) describes the massic activity of isotopes as a function of its radius (R). To further study the variation of $^{239+240}$Pu activities with particle radius, we did a linear fitting of Eq. (2) by least squares of the measured $^{239+240}$Pu activities as a function of the particle radius. As shown in Fig. 3, $^{239+240}$Pu activities decreased with the increasing particle radius as expected, and the fitted values for Pu isotopes correlated well with measured values ($R^2 = 0.748$). This result was consistent with previous result that most of the Pu isotopes were fixed to the soil as a surface coating (Lee et al., 2004).

It is noteworthy that the deviations between actual values and fitted results for the fractions exceeding 425 μm were markedly higher than that of smaller particles. The fitted values were almost twice the actual ones in the 600–1000 μm and 425–600 μm size fractions. When we omitted the large coarse fraction (> 425 μm) the correlation coefficient was improved to 0.932. This finding indicated that the spherical model gave better results in the small diameter range, especially less than 600 μm according to our study, which probably was due to the spherical approximation being more valid for smaller particles. For the large coarse fraction (> 425 μm), the spherical approximation could obtain larger surface area, thus, resulting in an overestimation of $^{239+240}$Pu activities by the model, which still need further investigations. Lee et al. (2004) studied Pu isotope distribution as a function of grain size using the same model. They obtained an extremely high correlation coefficient (0.994) for $^{239+240}$Pu, although the upper limit of the particle size fraction in their study was also 600 μm.

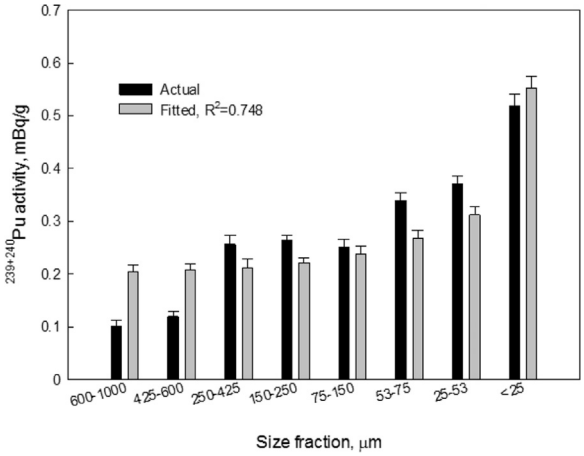

Fig. 3. $^{239+240}$Pu activities observed in various particle size fractions for the surface soil samples from central Inner Mongolia. Actual measured values are shown along with an estimated activity based on particle size.

3.3. Implications for the Asian Dust source identification

The long-term monitoring of anthropogenic radionuclides deposition in Japan conducted by the Meteorological Research Institute revealed that resuspension of the radioactivity from the land surface has been a major source in recent decades, leading to constant annual $^{239+240}$Pu depositions since 1985 (Igarashi et al., 2003; Hirose et al., 2008). Marked seasonal differences of $^{239+240}$Pu depositions, peaking in spring and being low in summer and fall, have been observed in Japan and Korea (Hirose et al., 2003, 2004). Hence, a hypothesis has been proposed that the temporal change of $^{239+240}$Pu deposition in eastern Asian is controlled by the long-range transport of suspended soil dust particles originating from East Asian arid and semi-arid areas as well as desert areas, i.e. Asian Dust events (Hirose et al., 2007). Recently, possible change of the Asian Dust source region, shifting from the arid zone to the desert-steppe zone undergoing desertification, have been suggested due to the higher $^{239+240}$Pu mass activities in depositions that occurred in the spring of the 2000s decade than those of the 1990s decade and this been supported by the changes of ^{137}Cs/^{90}Sr ratios (co-related to precipitation of the originating source) in the depositions of Japan (Igarashi et al., 2003, 2011). Hence, this change of the dust source region has been confirmed by the high $^{239+240}$Pu activities in Mongolian soil (Hirose et al., 2016).

Understanding the behavior of fine soil particles, especially particles sized smaller than 53 μm, is critical to describing the dust phenomenon because this fraction was considered as a candidate for the dust being suspended and transported by strong winds (Igarashi et al., 2011). No study of Pu isotopes giving detailed size fractions of soil particles in central Inner Mongolia (also close to the North China Plain) has been reported before. In this study, we analyzed $^{239+240}$Pu activities and ^{240}Pu/^{230}Pu atom ratios based on size fraction in surface soil samples. Observed $^{230+240}$Pu activity for particles < 53 μm was 0.414 ± 0.105 mBq/g and the ^{240}Pu/^{230}Pu atom ratio was 0.189 ± 0.019. These results were similar to those in the atmospheric depositions up to 0.4–0.5 mBq/g that occurred in the dust seasons of 2000, 2001, 2002, and 2006 (Igarashi et al., 2003), but slightly higher compared with the results of Liaodong Bay (average 0.361 mBq/g), in northeastern China, which is located on the dust transportation pathway from China to Japan (Xu et al., 2013). In this connection, Kurosaki and Mikami (2003) reported greater dust storm frequency from 2000 to 2003 than in the 1990s (1993–1999), and the area of frequent dust storms was expanded extensively from limited regions around southern Mongolia, the Badain Jaran Desert (located in the south-central part of Inner Mongolia as a section of the Gobi Desert) and the western Loess Plateau to the east, and also frequently occurred in regions around North China Plain, northeastern China and the Korean Peninsula. Therefore, our results support the assumption of new Asian Dust sources shifting from deserts and reasonably lead to the conclusion that the adjacent regions of North China Plain, i.e. central Inner Mongolia, are also potential Asian Dust sources in the 2000s, although further investigations into Pu isotope activities and characteristics in areas along dust transportation pathway, including the potential Asian Dust source regions, are necessary.

4. Conclusions

In the current study, we studied $^{239+240}$Pu activities in surface soil samples from central Inner Mongolia and we identified the possible source of Pu isotopes to be global fallout. The amounts of Pu isotopes adsorbed on fine particles were markedly higher than those of coarse particles due to more surface area provided for sorption, and this could be well-simulated by a spherical model for the particle diameters less than 600 μm. Also, $^{239+240}$Pu activities in fine particles smaller than 53 μm, which were recognized as the main portion of resuspended and long-term transportation dust, were comparable with those in adjacent regions and the depositions observed in Japan in the 2000s, indicating a possible new Asian Dust source region other than the Chinese deserts. Comprehensive studies of tracer isotopes in northern areas of China are expected to help clarify the above behaviors.

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