Recent trends of plutonium fallout observed in Japan: plutonium as a proxy for desertification

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Plutonium in monthly deposition samples collected in Tsukuba (the Meteorological Research Institute), Japan from 1990 to end of 2001 is reported, together with monthly plutonium deposition in Nagasaki and Yonaguni in 2000. The annual deposition of ^{239,240}Pu during the period from 1990 to 2001 shows no systematic inter-annual variation. However, monthly ^{239,240}Pu depositions show a typical seasonal variation with a maximum in spring season (March to April), which corresponds to seasonal cycle of soil dusts originating from the East Asian arid area. Plutonium isotopic ratios in the deposition samples suggest that significant amounts of the recent ^{239,240}Pu deposition observed in Japan are attributed to the resuspension of plutonium-bearing surface soil particles; resuspended plutonium originates from the East Asian arid areas. The recent increased tendency of ^{239,240}Pu content in residues in deposition samples may reflect desertification in the East Asian continent.

Introduction

Behavior of plutonium in the environment is still of concern because of its high toxicity and political reasons. Major sources of plutonium in the environment are global fallout from atmospheric nuclear weapons tests and accidental releases such as US satellite (SNAP 9A).[1] As a result of atmospheric nuclear explosions, about 15 PBq of ^{239,240}Pu and 0.3 PBq of ^{238}Pu have been globally released in the atmosphere,[2] whereas 1.3 PBq of ^{238}Pu was injected into the upper atmosphere from satellite burnup.[3] Local plutonium contamination has occurred at the crash-sites of aircraft,[2,4] around nuclear weapons facilities,[5,6] following the Chernobyl nuclear reactor accident[7] and discharge of nuclear wastes in the ocean.[8,9] Plutonium in atmospheric deposition around Japan until 1984 mainly originated from stratospheric fallout due to atmospheric nuclear detonations conducted by the US, the former USSR and China.[10]

In order to establish an environmental baseline of ^{239,240}Pu deposition, time series data of plutonium deposition should be observed. Since March 1957, our laboratory has continuously determined the monthly (partly quarterly) deposition of ^{239,240}Pu at the Meteorological Research Institute (MRI; in Tokyo until March 1980 and in Tsukuba since April 1980). The studies concerning plutonium fallout up to the end of 1980 have been reported by Miyake *et al.*[11–13] and Katsuragi *et al.*[14] It is important, using such comprehensive time series data sets, to elucidate factors controlling the past and present plutonium in fallout, including that arising from nuclear weapons testing, nuclear satellite burnup, the Chernobyl fallout and resuspension. The time series data are significant also from the view point of a better understanding of the long-term history of anthropogenic radionuclides in the environment.

Resuspension of deposited radionuclides is one of the largest current concerns in the field of environmental radioactivity. After the end of atmospheric nuclear weapons testing in 1980, as mentioned earlier, resuspension is considered to be the predominant mechanism for maintaining the small residual plutonium in the surface air and deposition.[15] Nicholson[16] has reviewed resuspension of radionuclides including Pu in contaminated areas, which are usually arid or semiarid regions. Atmospheric resuspension of radionuclides can be a secondary source of radioactive contamination after the cessation of primary sources such as global fallout and the Chernobyl release.[17,18] For studies on resuspension processes, it, implicitly, has been considered that the origin of resuspended anthropogenic radionuclides is due to local sources near sampling sites, *i.e.*, resuspension of surface soil particles.[15,19] On the other hand, Igarashi *et al.*[20,21] introduced the hypothesis that fallout ^{90}Sr and ^{137}Cs observed in Japan in the 1990s originated from resuspended radioactivity deposited on the East Asian continent arid zone, based on ^{137}Cs/^{90}Sr activity ratios and the ratios of activities to corresponding stable elements. Furthermore, desertification in the East Asian continent would enhance resuspension of radionuclide-bearing soil particles. This phenomenon would result in the increase of plutonium deposition in Japan as well as ^{90}Sr and ^{137}Cs.

In this paper, we describe the long-term monthly deposition data of plutonium observed at the MRI in Tsukuba during the period of January 1990 to end of 2001 together with plutonium deposition in Yonaguni and Nagasaki in 2000, and discuss the geochemical and meteorological factors controlling the plutonium deposition, based on the data of plutonium isotopes.

Sampling and measurements

Sampling of rainwater together with falling dust was carried out at the observation field of the Meteorological Research Institute (until the end of March, 1980 in Tokyo and thereafter in Tsukuba, about 60 km northeast of Tokyo), using an open surface collector with a surface area of 1 m^2. It has been confirmed that no serious discontinuity occurred in the record of the Pu deposition before and after the change of the sampling station.[14] Since April 1986, we have used a larger sampler with a surface area of 4 m^2 to determine extremely low-level anthropogenic radioactivity. In the year of 2000, additional sampling of monthly deposition was conducted in

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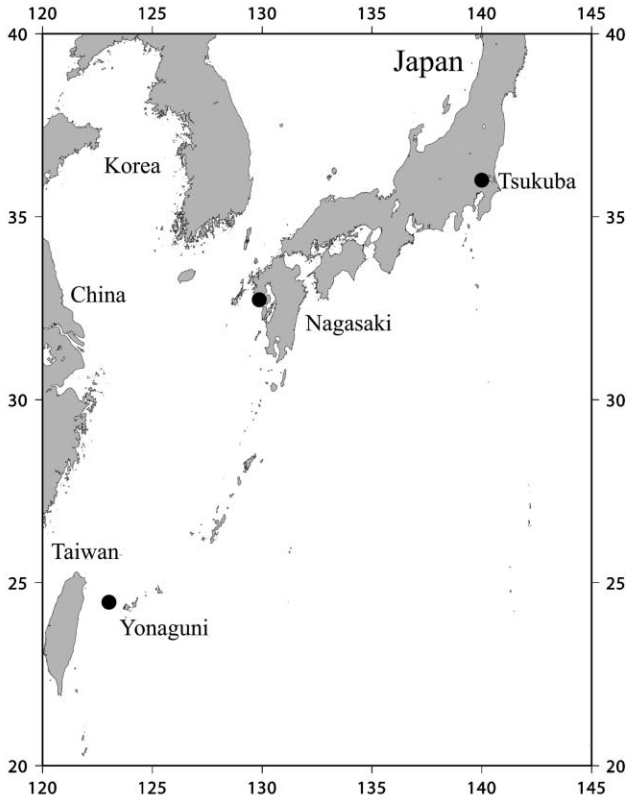

Fig. 1 Sampling locations of monthly ^{239,240}Pu deposition.

Nagasaki and Yonaguni Island (Fig. 1), using an open surface collector with a surface area of 3.69 m^2 (0.923 m^2 × 4).

A monthly composite of rainwater samples (10 to 1000 l) was evaporated to dryness in an evaporation dish or in a glass flask by using a rotary vacuum evaporator (Eyela NE-12). Monthly residue samples were weighed after drying in an oven at 110 °C and were transferred to a plastic container. The activity of γ-emitting nuclides in the residue samples was directly determined with a high-resolution intrinsic Ge γ-spectrometer. After the γ-ray measurement, the residue samples were digested with HNO_3 and the organic matter was decomposed. Finally, the residue was dissolved in 8 M HNO_3 solution. The acidic solution obtained was subjected to the radiochemical Pu separation procedure after addition of a known amount of ^{242}Pu to determine chemical yields.

The plutonium isotopes (^{238}Pu and ^{239,240}Pu) in monthly deposition samples were determined with α-spectrometry (Octête™ PC Alpha spectrometer with Canberra SSD of ⩽25 keV (FWHM)) after the chemical separation and purification using anion exchange resin (Dowex 1 × 8).[14,22] A detection limit of ^{239,240}Pu in monthly deposition samples is *ca.* 0.015 mBq m^{-2} under the present analytical conditions (counting time: 10^6 s). In order to determine precision and accuracy regarding plutonium analysis in deposition samples, reference materials of deposition samples were prepared as described in detail elsewhere.[22] The ^{239,240}Pu content in reference materials by our plutonium analysis procedure coincided with an average value of several institutes within less than 5%.[22] Measurements of ^{240}Pu/^{239}Pu atomic rations were carried out by high-resolution inductively coupled plasma-mass spectrometry (ICP-MS; PlasmaTrace 2, Micromass) described in detail elsewhere.[23] After the radiochemical separation (using anion exchange resin), a part of sample solution was injected into high-resolution ICP-MS system using an on-line purification and sequential injection technique including Sr-Spec and TEVA Spec resins. A sensitivity of high-resolution ICP-MS was 5.5 × 10^7 cps ppb^{-1} based on the measurement of ^{239}Pu. Background noises of peak areas of ^{239}Pu and ^{240}Pu were 3.3 and 1.3 cps, respectively.

Results

The monthly ^{239,240}Pu depositions observed in Tsukuba during the period from 1990 to end of 2001 are summarized in Table 1. The monthly ^{239,240}Pu depositions in the 1990s ranged from 0.04 ± 0.01 to 2.17 ± 0.16 mBq m^{-2}, which were the same orders of magnitude as those in the latter half of the 1980s. The monthly ^{239,240}Pu depositions observed in Nagasaki and Yonaguni from January 2000 to March 2001 are shown in Table 2. The level of the monthly ^{239,240}Pu depositions showed no difference among three stations.

Discussion

Time-series of ^{239,240}Pu deposition in Tsukuba

Fig. 2 shows time-series of annual ^{239,240}Pu deposition observed at the MRI in the period from 1957 to end of 2001. Temporal variations of the ^{239,240}Pu deposition in the period from 1957 to 1980 were controlled by the stratospheric fallout due to the atmospheric nuclear weapon testing, described in detail in our previous papers.[10,24] After the 26th Chinese nuclear test in October 1980, a marked increase of ^{239,240}Pu deposition occurred in the spring season in 1981. The annual ^{239,240}Pu deposition during the period from 1981 to 1984 decreased according to the stratospheric half-residence time of around 1 year.[24] A portion of stratospheric fallout ^{239,240}Pu in 1985, which is estimated from the stratospheric half-residence time of ^{239,240}Pu-bearing particles (1 year), is less than 20% of the total ^{239,240}Pu deposition. This finding suggests that the ^{239,240}Pu deposition in 1985 reached the background level in Japan. The present background level of the ^{239,240}Pu deposition is three orders of magnitude lower than that in the 1960s and more than three orders of magnitude greater than estimates by a model.[25] After 1985, the ^{239,240}Pu deposition seems to exhibit no systematic inter-annual variation, in contrast to temporal variation of the ^{239,240}Pu deposition in Munich, Germany, which showed a gradual decrease during the period from 1988 to 1998.[26] The annual ^{239,240}Pu depositions since 1999 tend to increase. The annual ^{239,240}Pu deposition in Tsukuba from 1985 to 2001 ranged from 1.7 ± 0.1 to 7.8 ± 0.3 mBq m^{-2}. The lowest annual ^{239,240}Pu deposition during this period occurred in 1989. The annual ^{239,240}Pu deposition in 1991 in Tsukuba was the same order of magnitude as that in Munich, Germany,[15] although the annual ^{239,240}Pu depositions in Munich in the latter half of the 1980s were slightly higher than that in Tsukuba, which may be due to the effect of the Chernobyl fallout. On the other hand, the annual ^{239,240}Pu deposition in Munich in the latter half of the 1990s was slightly lower than those in Tsukuba.[26] It must be noted that the highest annual ^{239,240}Pu deposition in Tsukuba during this period was observed in 2001. It, therefore, is necessary to understand factors controlling the variation of recent background ^{239,240}Pu in the deposition samples in Tsukuba.

Monthly ^{239,240}Pu deposition in Tsukuba during the period from 1985 to 2001 (Fig. 3) showed a typical seasonal variation with high values in spring and low in summer, even though there is no contribution of stratospheric fallout of ^{239,240}Pu, which shows spring maximum in the mid-latitude region as a result of active stratosphere-troposphere air exchange.[24] As a possible meteorological phenomenon related to the seasonal change of ^{239,240}Pu deposition, there is the long-range transport of surface soil particles originating from the Chinese deserts and arid areas. This seasonal change of atmospheric mineral dusts observed in Japan is related to arrival of continental dusts, so called "Kosa", from Chinese deserts and arid areas.

Table 1 Monthly ^{239,240}Pu deposition observed in Tsukuba during the period from 1990 to 2001 (unit: mBq m^{-2})[a]

	Jan	Feb	Mar	Apr	May	June	July	Aug	Sept	Oct	Nov	Dec
1990	0.27 ± 0.04	0.05 ± 0.01	0.66 ± 0.05	0.28 ± 0.04	0.09 ± 0.02	0.19 ± 0.04	0.04 ± 0.01	0.13 ± 0.03	0.11 ± 0.03	0.06 ± 0.02	0.10 ± 0.02	0.15 ± 0.03
1991	0.24 ± 0.06	0.26 ± 0.06	0.25 ± 0.06	0.46 ± 0.08	0.58 ± 0.12	0.42 ± 0.08	0.17 ± 0.04	0.09 ± 0.03	0.08 ± 0.03	0.17 ± 0.04	0.22 ± 0.06	0.04 ± 0.02
1992	0.31 ± 0.07	0.29 ± 0.05	0.72 ± 0.09	1.21 ± 0.10	0.44 ± 0.06	0.12 ± 0.03	0.16 ± 0.03	0.17 ± 0.03	0.47 ± 0.05	0.10 ± 0.03	0.08 ± 0.03	0.33 ± 0.08
1993	0.28 ± 0.07	0.35 ± 0.08	0.83 ± 0.14	2.10 ± 0.21	1.55 ± 0.23	1.27 ± 0.19	0.19 ± 0.05	0.16 ± 0.04	0.37 ± 0.07	0.25 ± 0.05	0.18 ± 0.05	0.22 ± 0.05
1994	0.36 ± 0.04	0.58 ± 0.08	0.41 ± 0.05	0.70 ± 0.07	0.68 ± 0.08	0.16 ± 0.04	0.23 ± 0.05	0.28 ± 0.05	0.11 ± 0.03	0.20 ± 0.04	0.25 ± 0.04	0.07 ± 0.03
1995	0.20 ± 0.04	0.30 ± 0.05	0.43 ± 0.06	0.45 ± 0.05	0.47 ± 0.07	0.13 ± 0.02	0.38 ± 0.11	0.13 ± 0.03	0.10 ± 0.03	0.23 ± 0.09	0.29 ± 0.03	0.13 ± 0.03
1996	0.31 ± 0.05	0.51 ± 0.06	0.78 ± 0.07	0.67 ± 0.06	0.42 ± 0.04	0.27 ± 0.04	0.15 ± 0.03	0.27 ± 0.04	0.12 ± 0.02	0.08 ± 0.02	0.13 ± 0.02	0.11 ± 0.02
1997	0.45 ± 0.06	1.4 ± 0.11	0.49 ± 0.07	0.38 ± 0.05	0.96 ± 0.11	0.20 ± 0.04	0.15 ± 0.06	0.15 ± 0.04	0.11 ± 0.03	0.15 ± 0.03	0.15 ± 0.03	0.10 ± 0.02
1998	0.11 ± 0.03	0.16 ± 0.03	0.97 ± 0.11	0.53 ± 0.07	0.18 ± 0.03	0.12 ± 0.02	0.07 ± 0.02	0.12 ± 0.02	0.12 ± 0.03	0.06 ± 0.01	0.10 ± 0.02	0.55 ± 0.05
1999	0.16 ± 0.02	0.63 ± 0.04	0.49 ± 0.03	0.45 ± 0.04	0.33 ± 0.04	0.19 ± 0.03	0.26 ± 0.04	0.21 ± 0.03	0.30 ± 0.05	0.21 ± 0.03	0.10 ± 0.023	0.14 ± 0.02

Table 2 Monthly ^{239,240}Pu deposition in Nagasaki and Yonaguni

	Nagasaki	Yonaguni
2000 Jan	0.32 ± 0.05	0.23 ± 0.04
Feb	0.52 ± 0.07	0.18 ± 0.03
Mar	2.53 ± 0.15	0.28 ± 0.03
Apr	1.01 ± 0.08	1.86 ± 0.09
May	1.50 ± 0.10	0.23 ± 0.04
June	0.24 ± 0.03	0.12 ± 0.02
July	0.20 ± 0.02	0.63 ± 0.09
Aug	0.13 ± 0.02	0.17 ± 0.03
Sept	0.18 ± 0.02	0.22 ± 0.03
Oct	0.09 ± 0.01	0.12 ± 0.02
Nov	0.15 ± 0.02	0.14 ± 0.02
Dec	0.27 ± 0.03	0.23 ± 0.03
2001 Jan	1.45 ± 0.11	0.48 ± 0.05
Feb	3.26 ± 0.14	0.51 ± 0.06
Mar	2.49 ± 0.13	0.74 ± 0.06

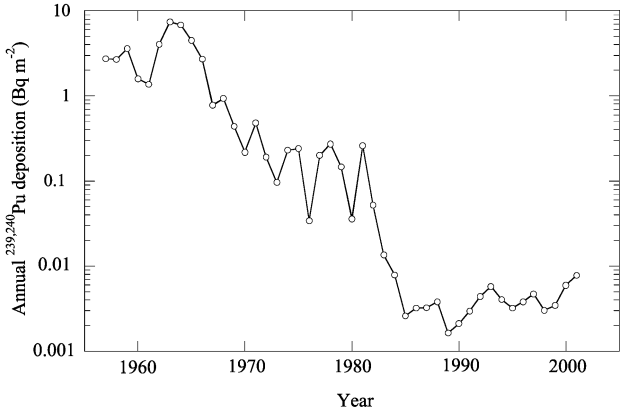

Fig. 2 Annual ^{239,240}Pu deposition observed at the MRI, Japan during the period from 1957 to 2001.

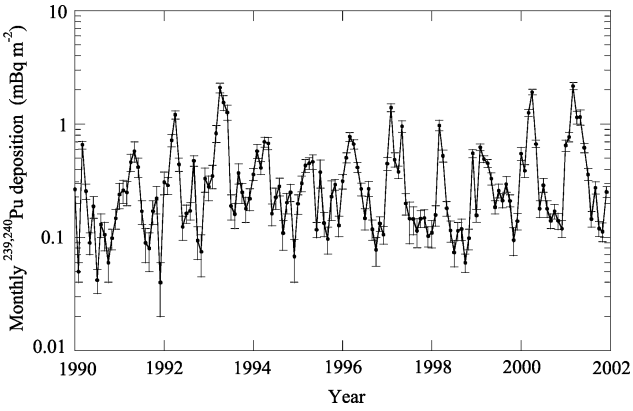

Fig. 3 Monthly ^{239,240}Pu deposition observed at the MRI during the period from 1990 to end of 2001.

Such "Kosa" events, which are marked in March to April in Japan, are large-scale dust storms over the continent behind the cold front accompanied with a low pressure. Igarashi *et al.*[20,21] revealed that ^{137}Cs/^{90}Sr activity ratios in deposition samples in Tsukuba (an average 2.1) preserved that of global fallout (1.6), and that ^{137}Cs/^{90}Sr ratios in Chinese desert soils maintain that of global fallout (an average: 2.1), whereas ^{137}Cs/^{90}Sr ratios in surface soil samples in Japan showed significantly higher values (an average 6.8). These findings lead the hypothesis that recent depositions of anthropogenic radionuclides observed in Japan are not mainly due to the local resuspension but long-range transport of Asian dusts.

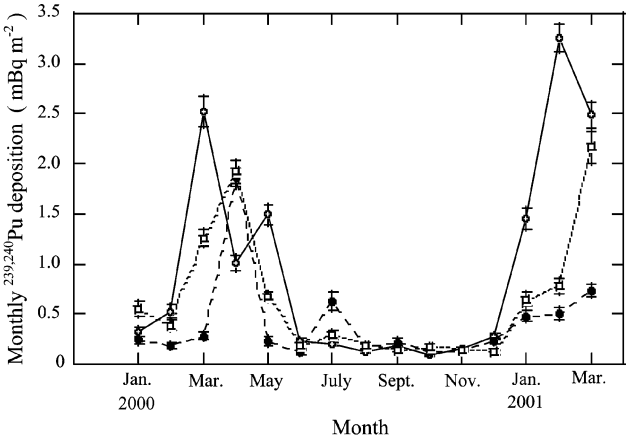

Fig. 4 Monthly ^{239,240}Pu deposition observed in Nagasaki, Yonaguni and Tsukuba in 2000. Open cross: Nagasaki, closed circle: Yonaguni, open square: Tsukuba.

Table 3 The annual ^{239,240}Pu deposition in Tsukuba, Nagasaki and Yonaguni in 2000

^{239,240}Pu deposition/mBq m^{-2}

Station	Annual sum	Dust period March–May	Non-dust period Jan, Feb, June–Dec
Nagasaki	7.13 ± 0.22	5.03 ± 0.20 (70%)[a]	2.10 ± 0.10
Tsukuba	6.01 ± 0.19	3.85 ± 0.15 (64%)[a]	2.16 ± 0.11
Yonaguni	4.42 ± 0.13	2.36 ± 0.10 (53%)[a]	2.06 ± 0.12

[a]The percentage of ^{239,240}Pu deposition in dust period related to the annual sum.

^{239,240}Pu deposition in Yonaguni and Nagasaki in 2000

Time series of monthly ^{239,240}Pu depositions in Yonaguni and Nagasaki from January 2000 to March 2001 together with those in Tsukuba is shown in Fig. 4. The monthly ^{239,240}Pu deposition in Yonaguni and Nagasaki in 2000 ranged from 0.12 ± 0.02 to 1.86 ± 0.09 mBq m^{-2} and 0.09 ± 0.01 to 2.53 ± 0.15 mBq m^{-2}, respectively. Maximum values of monthly ^{239,240}Pu deposition in 2000 were observed in March in Nagasaki and in April in Yonaguni. Occurrence of higher ^{239,240}Pu deposition corresponds to the dust season in the far Eastern Asia. A typical feature is that a highest ^{239,240}Pu deposition in Yonaguni in 2000 was observed only in April. In 2001, markedly high ^{239,240}Pu deposition in Nagasaki and Tsukuba occurred in early spring, whereas it is not remarkable in Yonaguni. On the other hand, monthly ^{239,240}Pu deposition in the non-dust period, except in July in Yonaguni, is the same level among three stations.

Annual ^{239,240}Pu deposition in 2000 is in the following order; Nagasaki > Tsukuba > Yonaguni. In order to understand the difference of the annual deposition between three stations, we calculated the cumulative ^{239,240}Pu deposition in the dust period (March–May) and the non-dust period (Jan, Feb, June–Dec), in which the dust period corresponded to the observation of the ''Kosa'' in Japan. The results are summarized in Table 3. A major part of ^{239,240}Pu in Japan was deposited in the dust season; *i.e.*, 70, 64, and 53% of annual ^{239,240}Pu deposition were deposited in the dust period in Nagasaki, Tsukuba and Yonaguni, respectively. On the other hand, there is no difference between sampling stations for the cumulative ^{239,240}Pu deposition in the non-dust period.

Isotopic ratios of ^{240}Pu/^{239}Pu

^{239}Pu and ^{240}Pu are major ones among plutonium isotopes, whose radioactive half-lives are 24,110 and 6,563 years, respectively. The atomic ratio of ^{240}Pu/^{239}Pu in fallout, which depends on the specific nuclear weapons design and test yield, is variable. The global fallout average ^{240}Pu/^{239}Pu atom ratio is 0.18, based on aerosols, soil samples and ice core data.[27–30] However, different nuclear test series can be characterized by either higher or lower ratios; *i.e.*, fallout from Nagasaki, Nevada test site and Semipalatinsk test site is characterized by generally lower ^{240}Pu/^{239}Pu ratios, 0.042, 0.035 as an average value, and 0.036, respectively,[31–34] whereas elevated ^{240}Pu/^{239}Pu ratios (0.21–0.36) have been measured in soil samples from Bikini atoll.[35–37] Therefore, ^{240}Pu/^{239}Pu ratios are a useful tool to identify sources of plutonium in the recent deposition samples.

^{240}Pu/^{239}Pu atomic ratios in the deposition samples collected in Tsukuba, Nagasaki and Yonaguni were determined by using ICP-MS. The results are shown in Table 4. The ^{240}Pu/^{239}Pu ratios in the monthly deposition samples in 1999 and 2000 in Tsukuba ranged from 0.16 ± 0.06 to 0.25 ± 0.01. There is no significant difference of the plutonium atomic ratios between deposition samples in the dust period and global fallout except March 1999. This finding confirms that most of the recent plutonium deposition in the dust period is originating from resuspended soil particles. Therefore, the result of plutonium isotopic ratios is consistent with the hypothesis that plutonium deposited in spring season in Japan is supported by aeolian transport of the continental soil dust. However, relatively high ^{240}Pu/^{239}Pu ratios in the deposition samples were observed in the non-dust period, whose causes are still future problems.

Table 4 ^{240}Pu/^{239}Pu atomic ratios deposition samples

	Tsukuba		Nagasaki	Yonaguni
	1999	2000	2000	2000
January	0.21 ± 0.02[a]	0.19 ± 0.03	0.22 ± 0.04	
February	0.18 ± 0.01	0.19 ± 0.02	0.17 ± 0.02	
March	0.25 ± 0.01	0.18 ± 0.01	0.19 ± 0.01	
April	0.19 ± 0.01	0.18 ± 0.01	0.19 ± 0.02	
May	0.21 ± 0.01	0.18 ± 0.01	0.17 ± 0.01	
June	0.22 ± 0.01	0.16 ± 0.06		
July	0.23 ± 0.01	0.18 ± 0.02	0.22 ± 0.01	0.18 ± 0.10
August	0.22 ± 0.01	0.21 ± 0.03	0.22 ± 0.02	0.17 ± 0.02
September	0.21 ± 0.01	0.20 ± 0.02	0.24 ± 0.04	0.17 ± 0.03
October	0.17 ± 0.02	0.22 ± 0.01	0.20 ± 0.03	0.23 ± 0.02
November	0.22 ± 0.02	0.20 ± 0.01	0.26 ± 0.02	0.18 ± 0.02
December		0.21 ± 0.02	0.19 ± 0.01	0.19 ± 0.01

[a]The standard deviation was calculated from 3 replicate Pu atomic ratio measurements.

^{239,240}Pu CIR as an index of resuspension

We introduce ^{239,240}Pu CIR as an index of resuspension, which is defined as ^{239,240}Pu contents in residue of monthly deposition samples, in order to elucidate contribution of resuspended ^{239,240}Pu, in which residual matter in deposition samples consists of surface soil, fly ash, sea salt and others. In the case when ^{239,240}Pu is newly introduced into atmosphere due to nuclear accidents and atmospheric nuclear explosions, the ^{239,240}Pu CIR is larger than the ^{239,240}Pu concentrations in surface soils, whereas it is smaller than that in the ^{239,240}Pu concentrations in surface soils when sea salts are occupied in significant part of residue in deposition samples because ^{239,240}Pu contents in sea salts, which is estimated to be less than 0.0003 mBq g^{-1} sea salts from ^{239,240}Pu concentrations in seawater,[38] is much lower than that in soils. The ^{239,240}Pu CIR during the period from 1990 to 2001 ranged from 0.021 $\pm$ 0.006 to 0.39 $\pm$ 0.03 mBq g^{-1}, which are the same order of magnitude as observed and estimated concentrations in soils (0.02 to 0.4 mBq g^{-1} in the Japanese surface soils,[39] 0.5 mBq g^{-1} in the northern China ($>45^{\circ}$ N), 0.2 mBq g^{-1} in the middle China (25°–45° N), and 0.1 mBq g^{-1} in the southern China ($<25^{\circ}$ N), which are estimated from ^{239,240}Pu/^{137}Cs ratio (0.02) in surface soil and ^{137}Cs concentrations in surface soils.[40] This finding suggests that there is no dominant source of ^{239,240}Pu except resuspension of surface soil particles.

The time series of the ^{239,240}Pu CIR in the period from 1990 to the end of 2001 is shown in Fig. 5. In order to elucidate the effect of the continental dust to the ^{239,240}Pu deposition, we divided the data into two seasons; one is the dust period from January to May including months potentially influenced by the transport of the continental dust and another non-dust period from June to December. The ^{239,240}Pu CIR in the dust period was higher than that in the non-dust period as shown in Fig. 5. Previous studies[10] reveal that the monthly ^{239,240}Pu deposition correlated to the weight of residue in the dust period, whereas there is no correlation between the monthly ^{239,240}Pu deposition and the monthly residue amount in the non-dust period. These findings suggest that significant amounts of resuspended ^{239,240}Pu, originating from the East Asian continent in the dust period, contributed to the ^{239,240}Pu deposition in Tsukuba.

Another typical feature of time-series of the ^{239,240}Pu CIR is that the ^{239,240}Pu CIR in the dust period increased during the period from 1990 to 2001, whereas there are no temporal changes of the ^{239,240}Pu CIR in the non-dust period. A median value of the ^{239,240}Pu CIR in the non-dust period is 0.06 mBq g^{-1}. In contrast to the temporal change of the ^{239,240}Pu CIR in Tsukuba, specific activities of ^{239,240}Pu in deposition samples in Munich, Germany, (around 0.1 mBq g^{-1}) corresponding to the ^{239,240}Pu CIR, showed no temporal change in the 1990s.[26] The mean increase rate of the ^{239,240}Pu CIR in the dust period is 0.015 mBq g^{-1} during the past 12 years. The increase tendency of the ^{239,240}Pu CIR in the dust period differs from that of recent radioactive deposition. Migration of ^{239,240}Pu in the soil layer leads the decrease of the ^{239,240}Pu concentrations in surface soils. On the other hand, the ^{239,240}Pu concentrations in surface soils depend on integrated amounts of radioactive deposition, as well as migration in the soil layer. The cumulative amount of ^{239,240}Pu deposition on the land surface due to global fallout is approximately proportional to the precipitation amount; relatively small accumulation of fallout ^{239,240}Pu occurred in deserts and arid areas, whereas higher soil ^{239,240}Pu appears in area with high precipitation in spring season of the 1960s and 1970s. The UNEP[41] reported that the desertified lands in China and Mongolia reached 70% of the total. Non-grassland currently formed due to over-cropping, in which surface soils contain relatively high ^{239,240}Pu, is a potential source of dust storms including relatively high ^{239,240}Pu. Therefore, the increase of the ^{239,240}Pu CIR in the dust period from 1990 to 2001 may reflect a signal of expansion of dust storms due to the desertification in the East Asian continent, especially in northern part of China.

^{239,240}Pu CIR in deposition samples in Nagasaki and Yonaguni were calculated using the weight of the corresponding dried residue. The ^{239,240}Pu CIR in Nagasaki and Yonaguni ranged from 0.05 $\pm$ 0.006 to 0.28 $\pm$ 0.02 mBq g^{-1}, and from 0.008 $\pm$ 0.002 to 0.33 $\pm$ 0.02 mBq g^{-1}, respectively. The ^{239,240}Pu CIR in Nagasaki, as in Tsukuba, was within the range of the current ^{239,240}Pu concentrations in surface soils. Temporal variations of the ^{239,240}Pu CIR in Nagasaki, Yonaguni and Tsukuba are shown in Fig. 6. The maximum ^{239,240}Pu CIR in three station occurred in April, whose concentrations (around 0.3 mBq g^{-1}) are similar to each other. The temporal variation of the ^{239,240}Pu in CIR in Nagasaki was similar to that in Tsukuba. On the other hand, the ^{239,240}Pu CIR in Yonaguni was lower than that in Nagasaki and Tsukuba, except April, May and July. This cause may be due to dilution effects by large sea salt depositions in Yonaguni.

Conclusion

The plutonium deposition at the MRI in Tsukuba, Japan has been reported until the end of 2001. Since 1985, the annual ^{239,240}Pu deposition, which ranged from 1.9 $\pm$ 0.1 to 7.8 $\pm$ 0.3 mBq m^{-2} y^{-1}, showed no systematic variation, although an increase tendency of the ^{239,240}Pu deposition was observed after 1999. The monthly ^{239,240}Pu deposition in Tsukuba, Nagasaki and Yonaguni showed a typical seasonal variation with high in spring and low in summer. Taken into account the meteorological

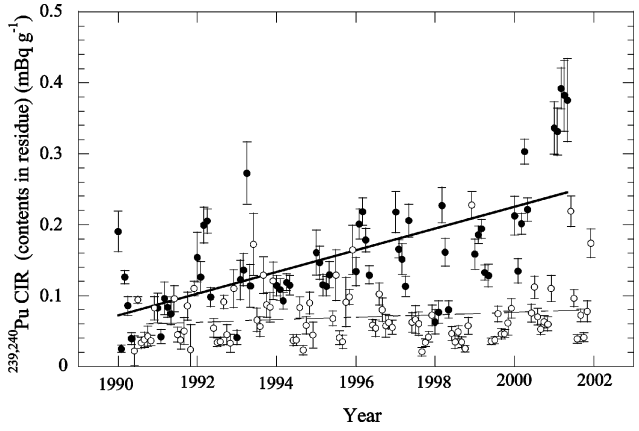

Fig. 5 ^{239,240}Pu CIR in monthly deposition samples during the period from 1990 to end of 2001. Closed and open circles show in dust period (Jan–May) and non-dust period (June–Dec), respectively.

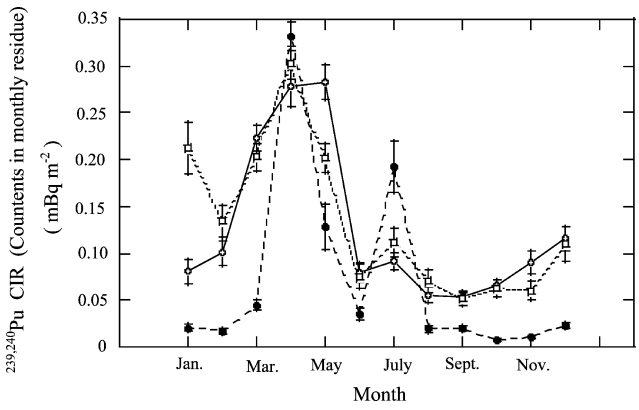

Fig. 6 ^{239,240}Pu CIR in monthly deposition samples in Nagasaki, Yonaguni and Tsukuba, in 2000. Open cross: Nagasaki, closed circle: Yonaguni, open square: Tsukuba.

conditions in Japan, the result suggests that the level of recent plutonium deposition in Japan is supported by long-range transport of plutonium-bearing soil particles, which is originating from dust storms occurring in the Chinese deserts and arid areas in spring season. The $^{240}Pu/^{239}Pu$ atomic ratios in deposition samples, which coincide with that in global fallout, are consistent with this hypothesis. We find out that the $^{239,240}Pu$ contents in residues in the dust period from 1990 to 2001 increased. This finding suggests that expansion of dust storms due to the desertification in the East Asian continent, especially northern part of China may result in an increase of the $^{239,240}Pu$ deposition in Japan.

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