

Atmospheric deposition of radioactive cesium (^{137}Cs) associated with dust events in East Asia

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Atmospheric deposition of radioactive cesium (^{137}Cs) associated with dust events in East Asia

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Synopsis

Since the cessation of atmospheric nuclear testing in 1980, there has been no known serious atmospheric contamination by radioactive cesium (^{137}Cs) apart from the Chernobyl nuclear reactor accident in 1986. There now remain only small amounts of anthropogenic radionuclides in the atmosphere that can be directly related to past testing. However, ^{137}Cs is still regularly found in atmospheric deposition samples in Japan. In this study, we analyzed ^{137}Cs monitoring data, meteorological data, and field survey results to investigate the recent transport and deposition of ^{137}Cs associated with dust phenomena.

Monthly records of nationwide ^{137}Cs deposition in Japan during the 1990s show a consistent seasonal variation, with higher levels of deposition occurring in spring. In March 2002, an unexpectedly high amount of ^{137}Cs was deposited in the northwestern coastal area of Japan at the same time as an Asian dust event was observed. Analysis of land-based weather data showed that sandstorms and other dust-raising phenomena also occurred in March 2002 over areas of Mongolia and northeastern China where grassland and shrubs predominated. Furthermore, radioactivity measurements showed ^{137}Cs enrichment in the surface layer of grassland soils in the areas affected by these sandstorms. These results suggest that grasslands are potential sources of ^{137}Cs -bearing dust. Continued desertification of the East Asian continent in response to recent climate change can be expected to result in an increase in ^{137}Cs -bearing soil particles in the atmosphere, and their subsequent re-deposition in Japan.

However, soil dust is also raised around Japanese monitoring sites by the strong winds that are common in Japan in spring, and this local dust might also contribute to ^{137}Cs deposition in Japan. To estimate the relative contributions of local and distant dust events to the total ^{137}Cs deposition, we monitored deposition of mineral particles and ^{137}Cs weekly in spring 2007. During one week of the period of weekly monitoring in 2007, we observed an Asian dust event. During that week, atmospheric ^{137}Cs deposition reached 62.3 mBq m^{-2} and accounted for 67% of the total deposition of ^{137}Cs during the entire eight-week monitoring period. This result suggests that the primary source of ^{137}Cs deposition in this part of Japan in spring is likely to be dust transported from the East Asian continent.

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Contents

Chapter I. General introduction	87
Chapter II. Recent atmospheric deposition of ¹³⁷ Cs in Japan and its relation to massive dust events	90
1. Introduction	90
2. Data and methods	90
(1) Monitoring data of ¹³⁷ Cs deposition, SPM, and meteorological factors	90
(2) Analysis of soil samples collected from Chinese grasslands	91
3. Results and discussion	92
(1) Deposition of ¹³⁷ Cs in Japan	92
(2) Dust - raising events on the East Asian continent in March 2002	95
(3) Radioactivity of grassland soils in northeastern China	96
4. Conclusions	100
Chapter III. Relative contributions of Asian dust and local dust to atmospheric deposition of ¹³⁷ Cs in Japan	101
1. Introduction	101
2. Data and methods	101
(1) Collection of atmospheric deposition samples	101
(2) Analysis of deposition samples	102
(3) Monitoring data of SPM and surface meteorological factors	102
3. Results and discussion	102
(1) Variations in the deposition of mineral particles and SPM concentrations	102
(2) Meteorological factors related to atmospheric deposition	105
(3) Variations in atmospheric ¹³⁷ Cs deposition and specific activity in relation to potential sources	106
4. Conclusions	107
Chapter IV. General features of atmospheric deposition of ¹³⁷ Cs-bearing Asian dust	108
1. Source area of ¹³⁷ Cs-bearing Asian dust	108
2. Relative contributions of dust phenomena to deposition of ¹³⁷ Cs in Japan	109
3. Contribution of ¹³⁷ Cs-bearing Asian dust to the atmospheric environment	109
Acknowledgements	110
References	110
Summary in Japanese	115

Chapter I

General introduction

According to the United Nations Scientific Committee on the Effects of Atomic Radiation (UNSCEAR, 2000a) ^{137}Cs is an anthropogenic radionuclide with a half-life of 30.1 years that is produced by nuclear explosions and accidental releases such as the Chernobyl nuclear reactor accident. Atmospheric testing of nuclear weapons from 1945 to 1980 is the primary source of environmental contamination by this radionuclide. Each test resulted in the uncontrolled release of substantial quantities of radioactive materials that were dispersed in the atmosphere and later deposited worldwide (UNSCEAR, 2000a). Since the cessation of atmospheric testing in 1980, there has been no known serious atmospheric contamination by ^{137}Cs apart from the Chernobyl nuclear reactor accident in 1986, and there now remain only small amounts of anthropogenic radionuclides in the stratosphere that can be directly related to past testing (Igarashi et al., 1996). Thus, the deposition of anthropogenic radionuclides such as ^{137}Cs from the stratosphere, the primary pathway for global radioactive fallout, has diminished greatly.

The anthropogenic radionuclides produced by atmospheric testing were transported mainly within the stratosphere and deposited gradually over wide areas (UNSCEAR, 2000a). Since the estimated stratospheric half-residence time of radioactive particles is about 1 year (Krey and Krajewsky, 1970; Katsuragi, 1983; Katsuragi and Aoyama, 1986; Hirose et al., 1987), by the 1990s, the ^{137}Cs component in the stratosphere presumably had decreased to negligible levels. Atmospheric ^{137}Cs from the Chernobyl nuclear accident is also believed to have declined rapidly after 1986, because most of it remained within the troposphere, where its half-residence time was less than 1 month (Aoyama et al., 1987; Aoyama, 1988; Hirose et al., 1990; Aoyama et al., 1991).

However, ^{137}Cs is still being detected in atmospheric deposition samples in Japan (Japan Chemical Analysis Center, 2003), indicating that ^{137}Cs has entered the troposphere from existing reservoirs and has subsequently been deposited in Japan as a direct consequence of some event other than the pre-1980 nuclear testing or the Chernobyl nuclear accident. Reservoirs of anthropogenic radionuclides may exist in the ocean or on land. Igarashi et al. (2003) studied the sea-salt transport of ^{137}Cs and the ^{137}Cs content of surface seawater and determined that the oceanic contribution to the annual deposition of ^{137}Cs on land is negligible, less than 0.2%. Thus, they assumed that the land surface is the major reservoir of ^{137}Cs .

The recently detected deposits of anthropogenic radionuclides apparently resulted from the re-suspension by wind uplift of soil particles contaminated by past nuclear testing, followed by re-deposition (Nicholson 1988; Igarashi et al., 1996; Rosner et al., 1997; Makhon'ko, 2000). Makhon'ko (2000) suggested that recent global ^{137}Cs contamination of the atmosphere near the ground is due primarily to anthropogenic activity, such as cultivation and vegetation burning (including forest fires), wind erosion, and duststorms. Because of their long half-life, anthropogenic radionuclides such as ^{137}Cs , ^{90}Sr , and Pu isotopes deposited on the ground during past testing have commonly remained in the soil, and re-suspension can apparently occur under windy conditions wherever the dry surface soil is exposed. Nicholson (1988) has reviewed the re-suspension of radionuclides in contaminated areas, which are usually in arid or semiarid regions. In general, previous studies of re-suspension processes have concluded that re-suspended anthropogenic radionuclides originate from local sources near observation sites (Rosner et al., 1997). Igarashi et al. (1996), however, hypothesized on the basis of $^{137}\text{Cs}/^{90}\text{Sr}$ activity ratios that ^{137}Cs and ^{90}Sr deposited in Japan in the 1990s originated not from local sources but from arid regions of the East Asian continent.

In continental East Asia, a considerable amount of soil dust is lifted into the troposphere in spring by frequent strong winds and sandstorms (Littmann, 1991; Parungo et al., 1994). In extreme cases, sandstorms can severely disrupt social and economic activities and even directly cause the loss of human life (Shao and Wang, 2003). Furthermore, large-scale suspension and transport of dust are increasingly attracting widespread scientific concern in relation to global climate change (Intergovernmental Panel on Climate Change [IPCC], 2001; Huebert et al., 2003; Mikami et al., 2006). Dust from

the East Asian continent is transported over long distances, as far as to the United States (Husar et al., 2001) and Europe (Grousset et al., 2003).

Dust particles in the atmosphere influence the global climate via their radiation forcing effects (Sokolik et al., 1998; Nakajima et al., 2003; Park et al., 2005; Aoki et al., 2005; Takemura et al., 2005). Moreover, temporal change in the occurrence of Asian dust events may reflect changing climate conditions. From 2000 to 2002, dust events on the Asian continent became significantly more numerous (Zhou and Zhang, 2003; Shao and Wang, 2003), and observations of dust in Japan simultaneously increased (Kurosaki and Mikami, 2003; Iino et al., 2004; Kanai et al., 2005). Furthermore, desertification in parts of the East Asian continent has increased the frequency of sandstorms and the resultant suspension of soil particles (Xu, 2006), which also would be expected to lead to increased ^{137}Cs deposition in Japan.

Peaks of ^{137}Cs deposition observed regularly at Japanese monitoring sites in spring have been attributed to Asian dust (Igarashi et al., 2003; Ueno et al., 2003). In particular, the frequency of ^{137}Cs deposition increased remarkably in the 2000s (Fig. 1), and showed a positive linear correlation with the frequency of Asian dust events (Fig. 2), suggesting that the ^{137}Cs originated from continental sources. Atmospheric deposition in Japan is likely to be composed of both Asian dust and dust locally suspended near the observation sites, but Igarashi et al. (2005) suggested that contributions from aeolian dust sources such as the Saharan desert were also possible. Because France conducted nuclear weapons tests from 1960 to 1966 in the Saharan desert, this area is also an important potential source of radionuclides such as ^{137}Cs (Papastefanou et al., 2001; Hernández et al., 2005a; Hernández et al., 2005b). However, Igarashi et al. (2005) were unable to demonstrate a contribution of Saharan dust to ^{137}Cs deposition in Japan.

Short-lived anthropogenic radionuclides such as ^{95}Zr and ^{140}Ba were the main contributors to external exposure of humans to radioactivity during active nuclear testing. However, ^{137}Cs became the most important contributor in the 1960s, and at present it is the only radionuclide contributing to continuing external exposure from deposited anthropogenic radionuclides (UNSCEAR, 2000a). Although current monthly ^{137}Cs deposition levels in Japan are less than 1 Bq m^{-2} (Japan Chemical Analysis Center, 2003) and seem to have little effect on human health, the long-term effect of human exposure to ^{137}Cs is not clear. To predict future deposition of ^{137}Cs in Japan and to evaluate its possible effects on human health, it is essential to investigate both possible source areas and transport processes. Therefore, in this study, we analyzed ^{137}Cs monitoring data, meteorological data, and field survey results and examined the transport and deposition of ^{137}Cs associated with recent dust phenomena. In chapter II, we describe the transport of ^{137}Cs in relation to massive Asian dust events, and the primary sources of ^{137}Cs -bearing dust, determined from surface meteorological data and radioactivity measurements of continental soils. In chapter III, we describe the relative contributions of Asian dust and local dust to springtime ^{137}Cs deposition in Japan, estimated from weekly observations. Finally, in chapter IV, we sum up our results in a general discussion of the atmospheric deposition of ^{137}Cs -bearing Asian dust in order to clarify the causes of recent increases in ^{137}Cs deposition in Japan.

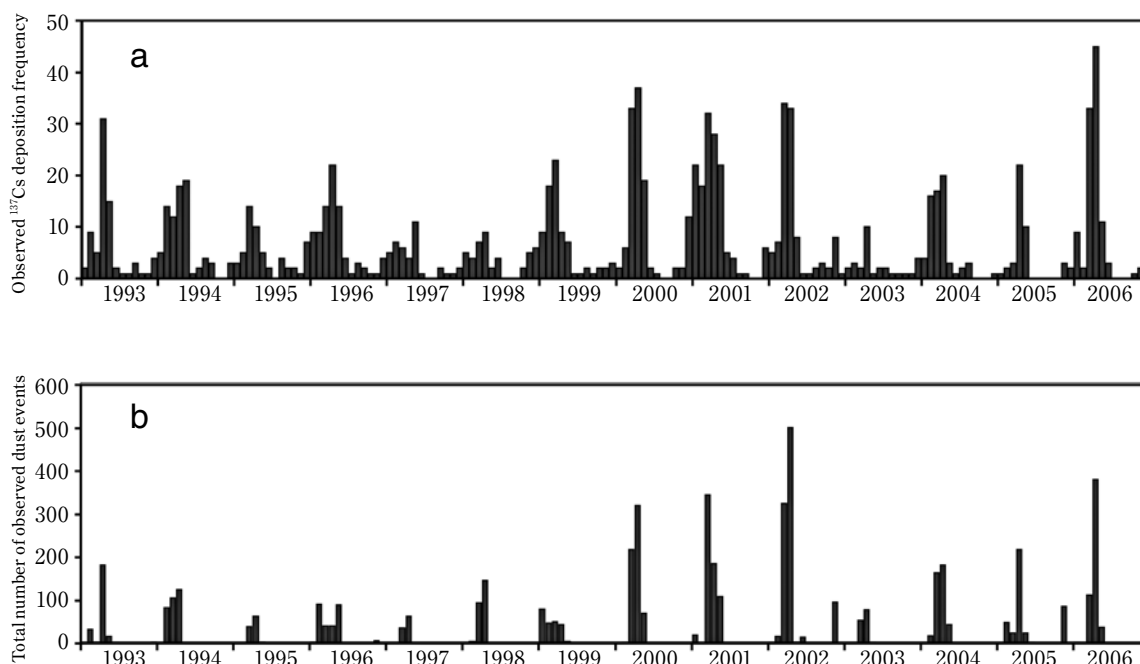


Fig. 1. Monthly frequency distributions of ^{137}Cs deposition (a) and observed dust events (b) in Japan. The ^{137}Cs deposition frequency is defined as the number of monitoring stations observing significant ^{137}Cs deposition each month at 47 sites in Japan, as determined from radiochemical analysis data in the MEXT database (Japan Chemical Analysis Center, 2009). Total number of observed dust events is defined as total number of days when 85 stations observe Asian dust for one month (Japan Meteorological Agency, 2008).

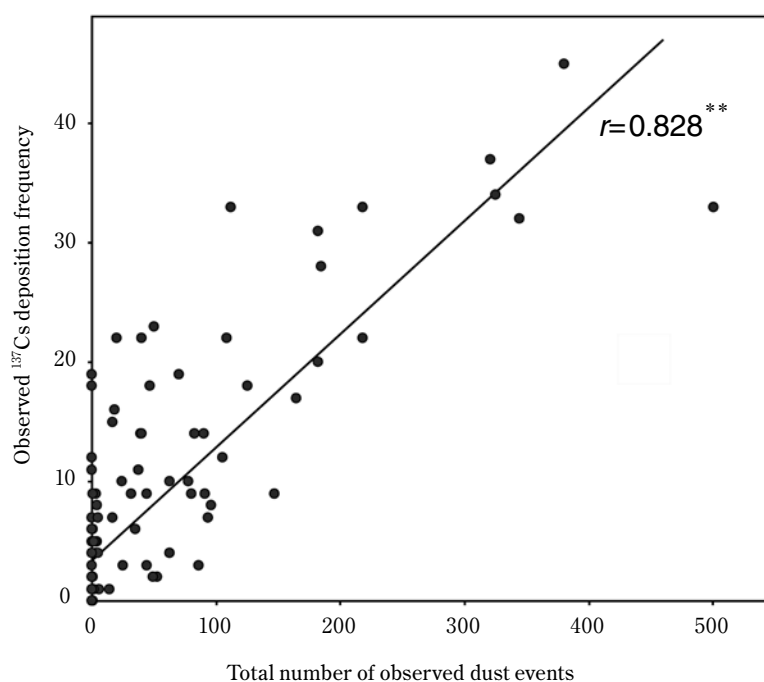


Fig. 2. Relationship between monthly ^{137}Cs deposition frequency at 47 sites and monthly observed dust events at 85 sites from 1993 to 2006 ($n = 168$).

Chapter II

Recent atmospheric deposition of ^{137}Cs in Japan and its relation to massive dust events

1. Introduction

Recent levels of ambient anthropogenic radionuclides must be understood in order to predict future deposition and evaluate their possible effects on human health. In Japan, the Ministry of Education, Culture, Sports, Science and Technology (MEXT) has conducted monthly observations of ^{137}Cs deposition in 47 Japanese prefectures since the late 1950s. Because only a few Russian and German publications have reported continuous observation results of ^{137}Cs deposition in other countries (Gritchenko et al., 2001; Rosner and Winkler, 2001), the monitoring data provided by MEXT are particularly valuable for investigating the atmospheric transport and deposition of ^{137}Cs . Our objective in this chapter is to characterize recent ^{137}Cs deposition in Japan in relation to Asian dust events by using the MEXT data, and to report the primary source of ^{137}Cs -bearing dust as determined from surface meteorological data of East Asia and radioactivity measurements of soil samples.

2. Data and methods

(1) Monitoring data of ^{137}Cs deposition, SPM, and meteorological factors

We obtained nationwide data of ^{137}Cs deposition in Japan from the radiation database maintained by MEXT (Japan Chemical Analysis Center, 2009). Then, using radiochemical analysis data from the database, we examined monthly variations of ^{137}Cs deposition in Japan. Furthermore, we obtained suspended particulate matter (SPM) concentration data and surface meteorological data for Japan from the Environmental Information Center (EIC) of Japan and the Japan Meteorological Agency (JMA) and investigated the relationship between ^{137}Cs deposition and meteorological factors.

SPM, which is used as the national ambient air quality standard in Japan, is defined as airborne particles with a diameter of 10 μm or less. PM10 is also used extensively in air quality monitoring. The difference between SPM and PM10 is that PM10 includes all particles that pass through a size-selective inlet with a 50% cut-off at the aerodynamic diameter of 10 μm , whereas SPM has a 100% cut-off at the same diameter.

We determined and mapped the frequency of sandstorms and other dust-raising phenomena in East Asia from surface meteorological data. We obtained six-hourly (four times a day) meteorological data based on surface synoptic observation (SYNOP) reports for East Asia from the JMA. SYNOP is a numerical code used for reporting weather observations, and SYNOP reports are typically sent worldwide every three or six hours from land-based weather stations. The code numbers 07, 08, 09, 30-35, and 98 in current SYNOP weather records indicate dust-raising events, such as sandstorms and sand whirls. We excluded code number 06 from our study because it refers to dust already suspended in the air, rather than dust raised by wind in the vicinity of the weather station. We defined the dust outbreak frequency as the number of sandstorms and other dust-raising phenomena as a percentage of the total number of weather observations (Kurosaki and Mikami, 2003) made during a month at each station. However, at a considerable number of stations, the records were incomplete. Because monthly frequencies calculated from only few observation records would be unreliable, we included only data sets with a threshold value of 100 observations per month in our study.

To identify the type of land cover in areas where dust-raising phenomena were frequently observed, we obtained Global Ecosystems data (Global Land Cover Characteristics Version 2) from the United States Geological Survey (United States Geological Survey, 2009). For this study, we categorized the Global Ecosystems land cover types into seven groups (Table I).

In addition, we obtained processed precipitation data for 1961 to 1990 for the East Asian continent from the JMA to investigate the relationship between precipitation and soil radioactivity in the area where dust-raising events were frequently observed during those years. We used these data to map mean annual precipitation by the minimum curvature interpolation method.

Table 1. Land cover groups used in this study, derived from US Geological Survey Global Land Cover Characteristics Version 2 (GLCC Ver. 2).

Defined land cover groups	Original codes in GLCC global ecosystems legend *
Forest and woods	4, 5, 21-24, 26, 27, 29, 33, 34, 43, 60-62, 90, and 91
Grassland	2, 10, 40, and 42
Shrubs	16, 17, 51, and 64
Grass, Shrubs, and crops	94
Cropland	19, 30, 31, 36-38, 55-58, 93, and 96
Desert	8, 11, and 50
Others	1, 9, 12, 14, 44, 45, 53, and 63

* Although 96 ecosystem types, based on their land cover mosaic, floristic properties, climate, and physiognomy, are defined in the original data set, for this study we clustered these 96 types into seven groups.

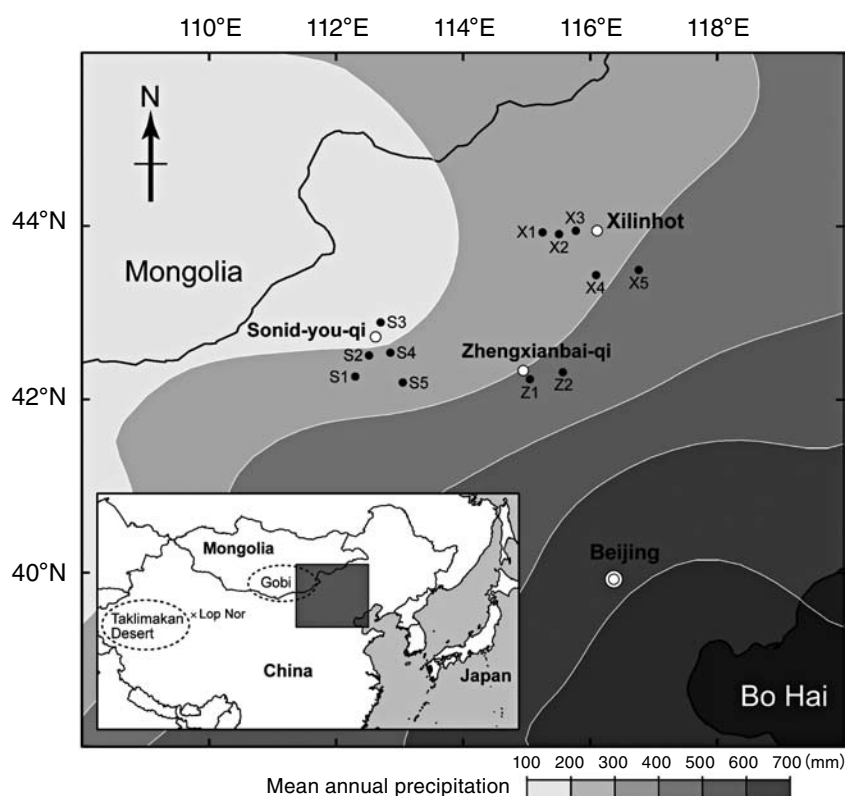


Fig. 3. Soil sampling sites in northeastern China.

Black circles with site number prefixes S, X, and Z indicate samples from Sonid-you-qi ($42^{\circ}45' \text{N}$, $112^{\circ}39' \text{E}$), Xilinhot ($43^{\circ}57' \text{N}$, $116^{\circ}05' \text{E}$), and Zhengxianbai-qi ($42^{\circ}18' \text{N}$, $115^{\circ}00' \text{E}$), respectively. Contours show annual precipitation interpolated by the minimum curvature method from sites of known precipitation based on data obtained from the JMA.

(2) Analysis of soil samples collected from Chinese grasslands

We carried out field surveys each summer from 2001 to 2003 in the central part of the Inner Mongolia Autonomous Region of China, where desertification has become a problem. We collected 2-cm-thick soil samples down to a depth of 20 cm from three semiarid grassland areas (Fig. 3) and determined the vertical profiles of ^{137}Cs in the soil.

Each sample was air-dried and passed through a 2-mm-mesh sieve to remove pebbles and plant roots, and then

put into a plastic container for a gamma spectrometric assay. The ^{137}Cs activities were measured by counting 662-keV gamma-ray emissions with germanium detectors (Canberra XtRa). The detection limit was defined as 3σ of the counted value (Cooper, 1971) and was estimated to be around 70 mBq. This value is likely inflated by increased background counts due to Compton scattering of gamma-rays from ^{40}K in the soil samples. The standard date used for decay correction was 1 January 2002. We measured ^{137}Cs activity in mass-based units of mBq g^{-1} . We calculated total activity (the ^{137}Cs inventory, expressed in area-based units of Bq m^{-2}) as follows:

$$^{137}\text{Cs inventory} = \sum_{i=1}^n 10^3 C_i B_d D_i$$

where i is the sampling depth and n is total number of samples with detectable ^{137}Cs , C_i is ^{137}Cs activity for sample i (Bq kg^{-1}), B_d is the bulk density (t m^{-3}) of topsoil, and D_i is the depth (m) for sample i (Walling and Quine, 1993; Yan and Shi, 2004). To investigate the relationship between the vertical ^{137}Cs profile and soil characteristics, we measured the contents of fine mineral particles and organic matter in the soil samples. We determined clay (particles smaller than $2 \mu\text{m}$) and silt ($2\text{--}20 \mu\text{m}$) contents of surface soil samples by the pipette method (Gee and Bauder, 1986), and the soil organic matter content by the loss-on-ignition method (Nelson and Sommers, 1996).

3. Results and discussion

(1) Deposition of ^{137}Cs in Japan

A great deal of ^{137}Cs was released by atmospheric nuclear testing in the period leading up to 1980 (UNSCEAR, 2000a). Residual ^{137}Cs in the atmosphere was continuously deposited in Japan throughout the 1980s (Fig. 4). In addition, the Chernobyl nuclear reactor accident in April 1986 caused intense deposition of ^{137}Cs over Japan, but this level of deposition was not sustained. In the 1990s, some deposition of ^{137}Cs was observed each spring, apparently controlled by re-suspension processes on the East Asian continent. Unexpectedly high levels of ^{137}Cs deposition were observed in March 2002, and the highest levels since 1986 were observed at several sites in the northwestern coastal area of Japan (Fig. 5). For example, the highest level recorded in Japan in March 2002 was 0.82 Bq m^{-2} at Aomori. Since no nuclear explosions or serious accidents involving the release of ^{137}Cs occurred in the early 2000s, an increased contribution from suspended dust appears to be the only possible explanation for this phenomenon.

A massive sandstorm on the East Asian continent on 19 and 20 March 2002 was reported to be the largest sandstorm in that region since the 1990s (Jin and Yan, 2003). The Korean Peninsula and the Japanese Islands were seriously affected by transported dust a few days later (Chung et al., 2003). Figure 6 shows the daily average concentrations of SPM and daily rainfall observed in Japan in March 2002. A clear SPM concentration peak was observed on 22 March at Sapporo, Niigata, and Matsue in the northwestern coastal area of Japan, where considerable ^{137}Cs deposition was recorded in March 2002. In contrast, no apparent SPM peaks were observed at Sendai, Tsukuba, or Nagoya in the southeastern coastal area of Japan, where little deposition of ^{137}Cs was recorded. Thus, we inferred that the high dust concentrations and the high depositions of ^{137}Cs observed mainly in the northwestern coastal area at this time were from dust transported from the East Asian continent. However, strong winds associated with the passing of cold fronts can also cause suspension of locally derived dust over Japan. As the dust event in March 2002 was accompanied by a cold front and its associated storm activity (Iino et al., 2004), we investigated the possible contribution of locally suspended dust.

At Sapporo, Niigata, and Matsue, frequent rainfall or snowfall was observed during the days before the sharp increase in SPM (Fig. 6). The total precipitation recorded in March at Sapporo, Niigata, and Matsue was 45, 135.5, and 160.5 mm, respectively. In addition, at Sapporo, snow covered the ground continuously from 1 to 26 March. Thus, we inferred that the amount of local dust raised by wind uplift in these areas was small during March 2002 because of the wet soil conditions and snow cover.

Figure 7 shows the hourly SPM concentrations and hourly wind speed in Japan during the dust event from 21 to 23 March 2002. The SPM peak observed in the northwestern coastal area of Japan at Sapporo, Niigata, and Matsue was not

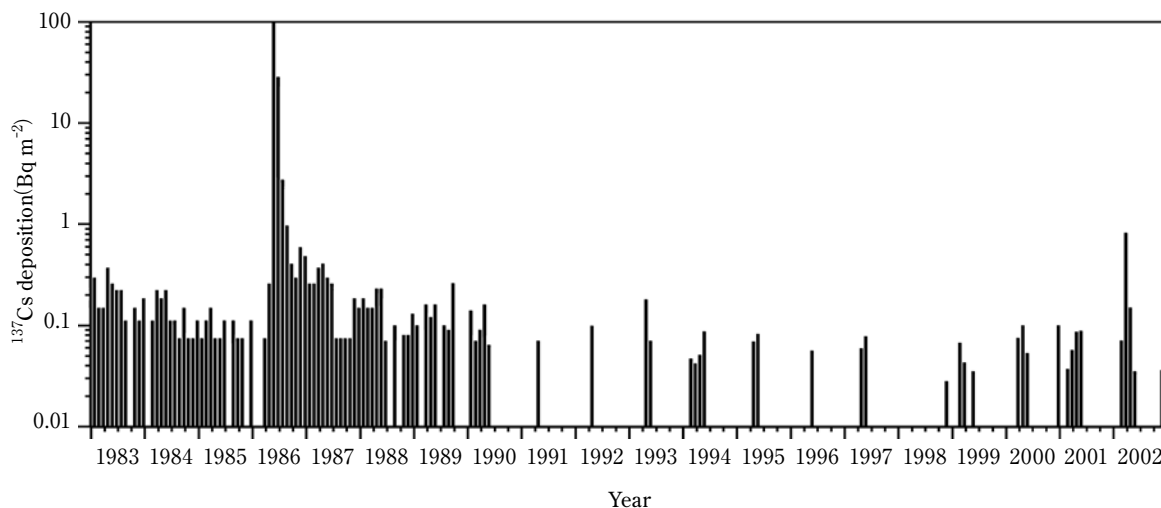


Fig. 4. Monthly atmospheric deposition of ^{137}Cs from 1983 to 2002 at Aomori, Japan. Monthly ^{137}Cs deposition data were obtained from the MEXT database (Japan Chemical Analysis Center, 2009). The sudden increase in 1986 reflects fallout from the Chernobyl reactor accident on 26 April 1986. In May 1986, atmospheric deposition of ^{137}Cs reached 99.9 Bq m^{-2} , more than 2 orders of magnitude higher than that of the previous month. In March 2002, the monthly ^{137}Cs deposition was 0.82 Bq m^{-2} , the highest level recorded since 1986.

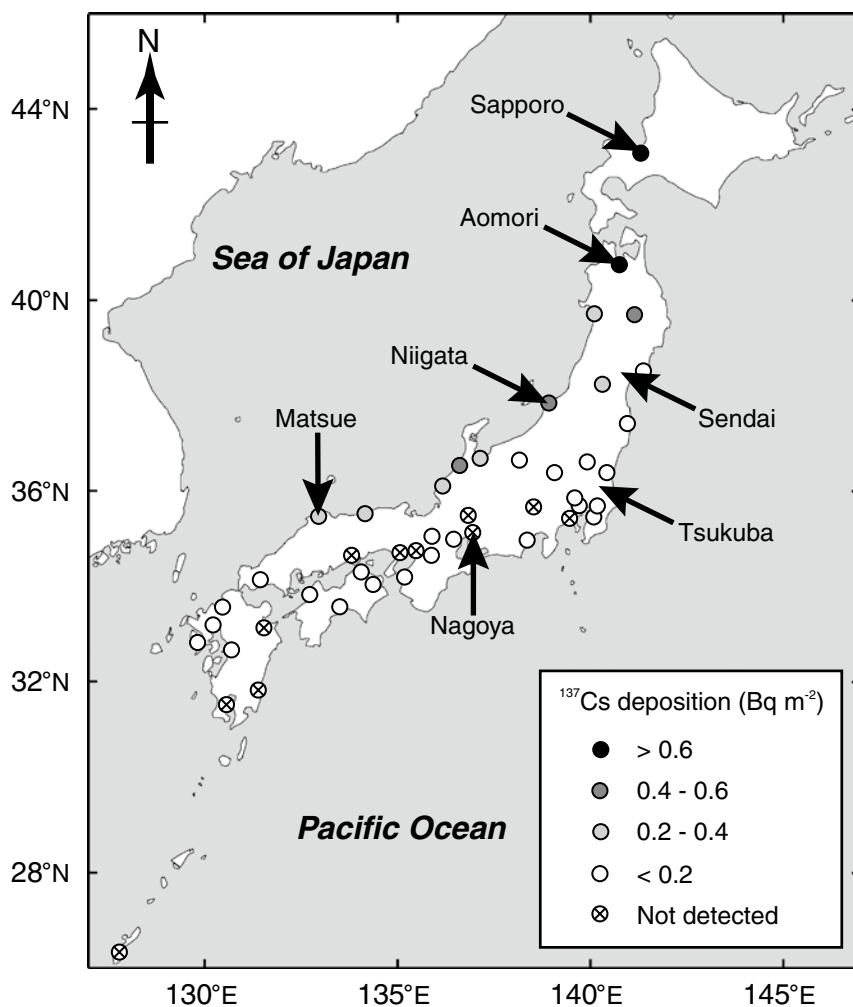


Fig. 5. Deposition of ^{137}Cs over Japan in March 2002. Monthly ^{137}Cs deposition data were obtained from the MEXT database (Japan Chemical Analysis Center, 2009).

coincident with the peak of wind speed, which was caused by the passing cold front. At Niigata, the SPM peak lagged the wind speed peak by five hours and at Matsue, the lag was 17 hours. These observations suggest that the increases in SPM and ^{137}Cs deposition along the northwestern coast of Japan were caused by the inflow of an air mass containing suspended dust after the cold front had passed. Although strong winds were observed as the cold front passed, no apparent SPM peak was observed at Sendai or Tsukuba in the southeastern coastal area of Japan. Thus, the contribution of locally suspended dust to the deposition of ^{137}Cs during the Japanese dust event of March 2002 was probably low.

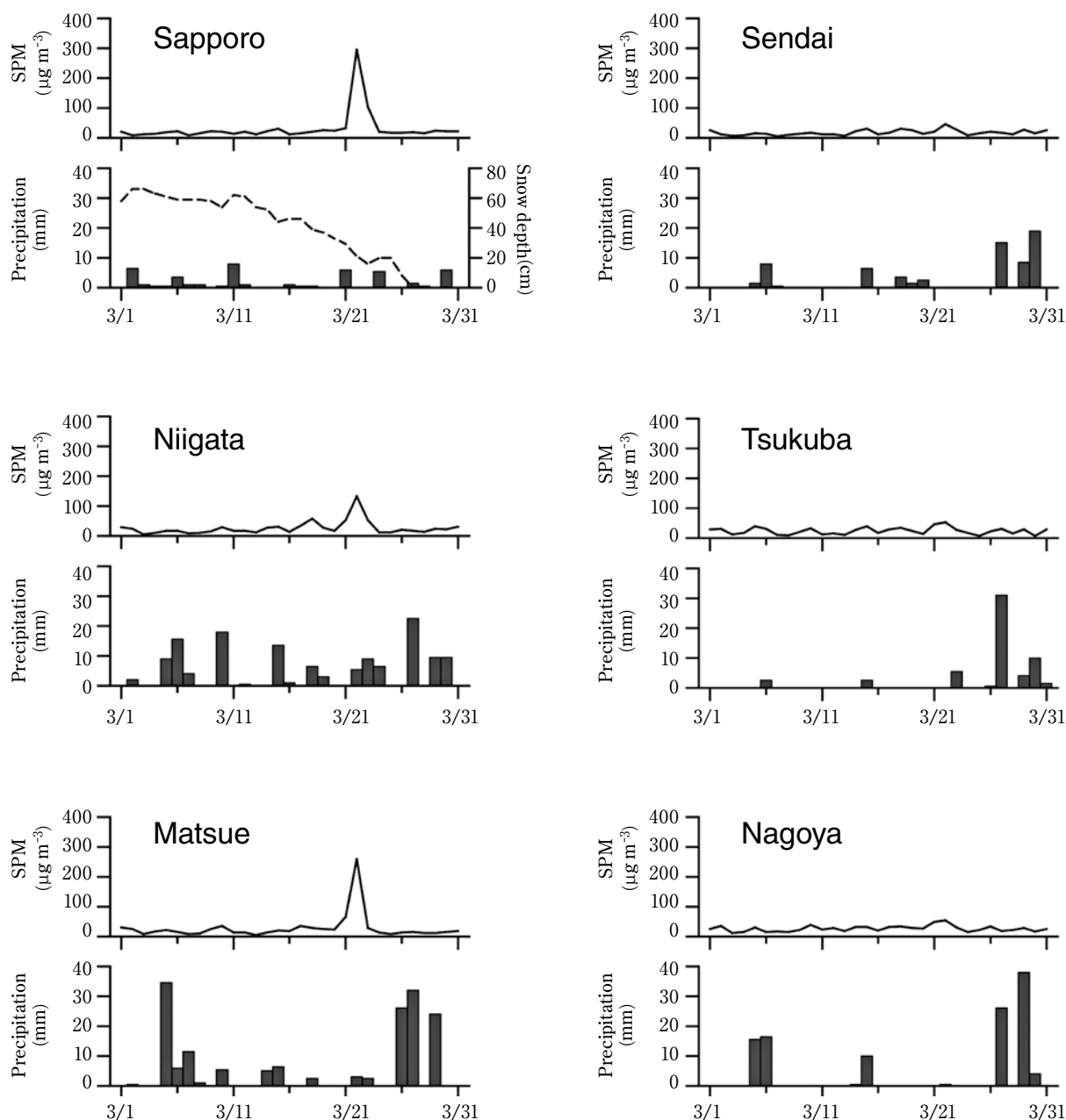


Fig. 6. Daily average SPM concentration and daily precipitation in March 2002.

Snow depth is also shown for Sapporo (broken line). The locations of these six observation stations are shown on Figure 5. Daily average SPM concentrations were calculated from hourly data provided by the EIC. Daily precipitation and snow depth data were obtained from public records of the JMA.

(2) Dust-raising events on the East Asian continent in March 2002

The arid areas of western China have been regarded as the major source of Asian dust (Xuan and Sokolik, 2002). Igarashi et al. (1996) suggested that recent ^{137}Cs deposition in Japan originated from dust raised in western China, which is also where nuclear testing was previously carried out. Between 1964 and 1980, 22 atmospheric tests were conducted at Lop Nor (40°N , 90°E) in western China (UNSCEAR, 2000a). However, as shown in Figure 8, in March 2002 sandstorms and other dust-raising phenomena were observed most frequently over Mongolia and northeastern China, which are far

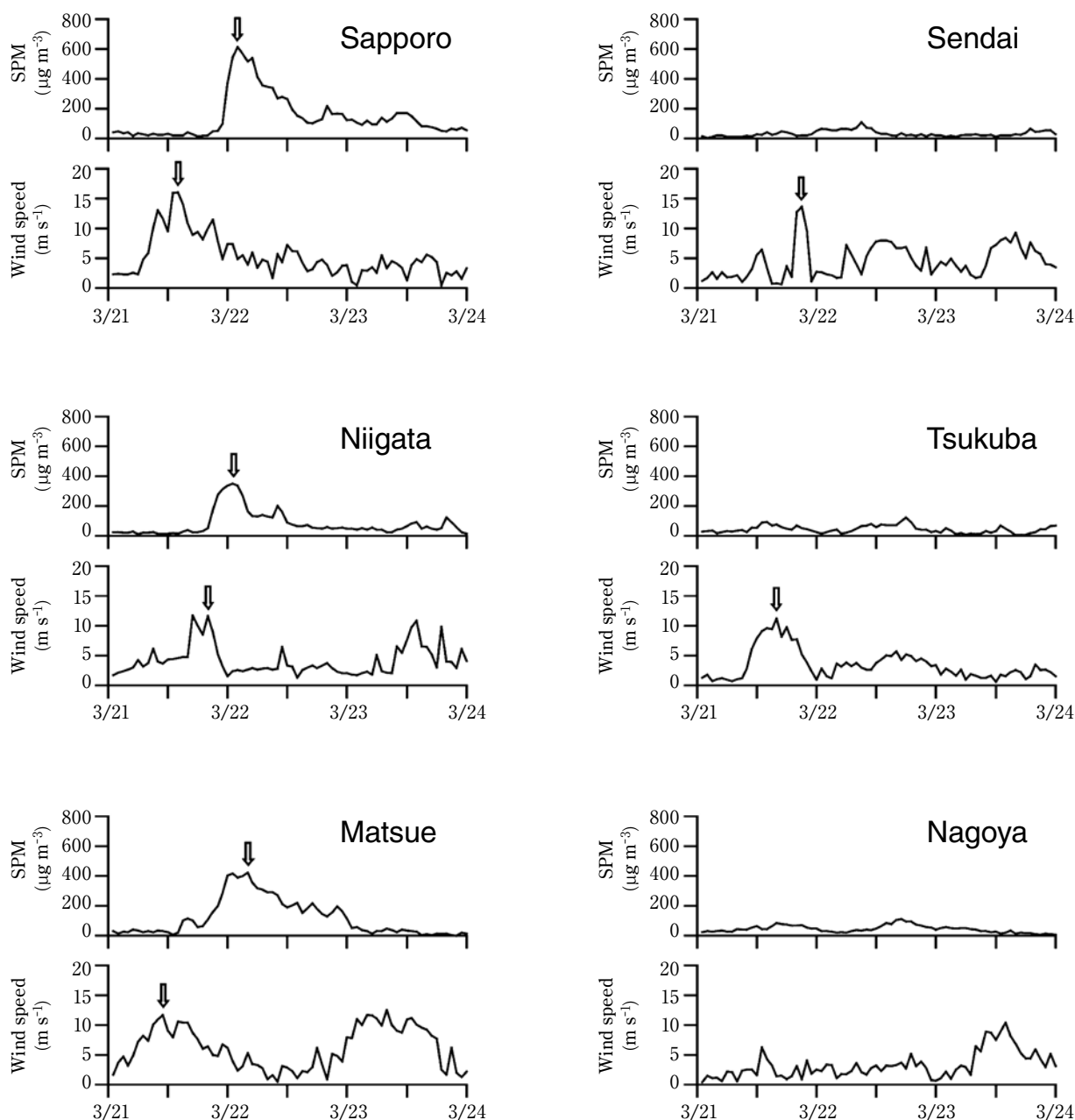


Fig. 7. Hourly SPM concentrations and hourly wind speed during the dust event from 21 to 23 March 2002.

The hourly SPM concentrations and the hourly wind speed data were obtained from the EIC and public records of the JMA, respectively. Arrows indicate SPM and wind speed peaks during the monitoring period. SPM peaks were observed at Sapporo, Niigata, and Matsue, which are in the northwestern coastal area of Japan. These peaks did not coincide with the peaks of wind speed, which were caused by a passing cold front. The lag time of the SPM peak after the wind speed peak was five hours at Niigata and 17 hours at Matsue. No apparent SPM peaks were observed at Sendai, Tsukuba, or Nagoya, which are in the southeastern coastal area of Japan.

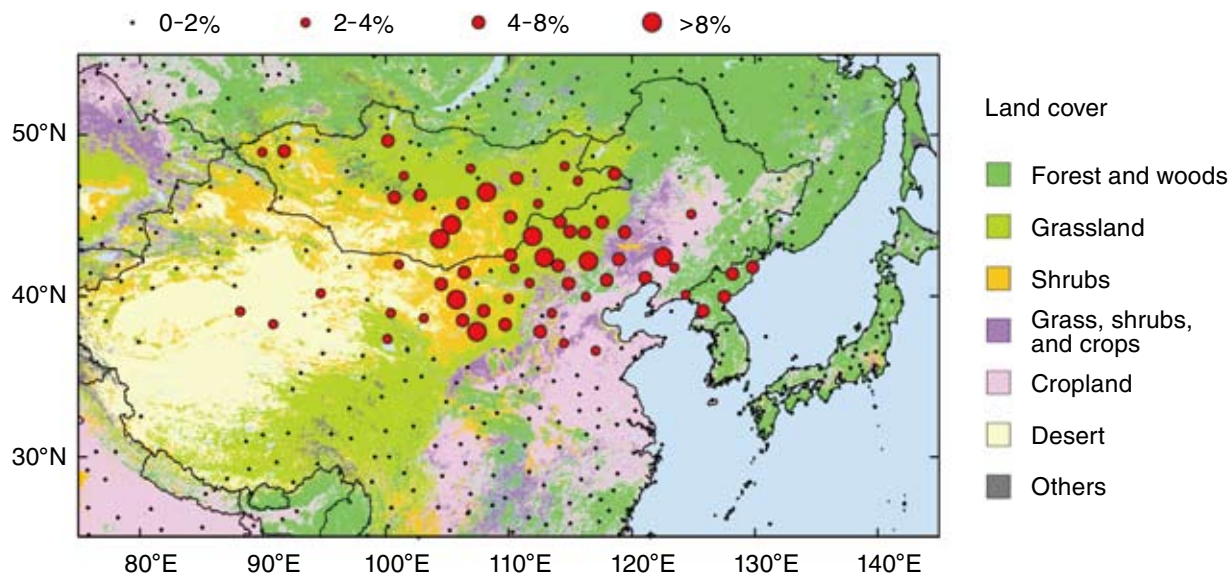


Fig. 8. Dust outbreak frequencies in East Asia in March 2002.

Circles indicate the frequencies calculated from SYNOP weather records, with larger circles indicating higher frequencies. Thick solid lines indicate country boundaries. Colors represent the land cover types (Table 2) derived from US Geological Survey Global Land Cover Characteristics.

from the nuclear test site. It is reasonable to infer that the massive sandstorm of 19 to 20 March 2002 contributed greatly to the observed high frequency of dust-raising events that month. Park and In (2003) determined the spatial distribution of dust over East Asia between 20 and 23 March 2002 by using aerosol index data obtained by the satellite-borne Total Ozone Mapping Spectrometer (TOMS). They reported that high values of the index over northeastern China on 20 March coincided with dust-raising events observed at land-based weather stations. The aerosol layer combined with new dust raised in northeastern China and moved gradually southeastward, producing a very thick dust layer over the Korean Peninsula on 21 March. Thereafter, the dust layer moved progressively eastward to northern Japan.

It thus appears that the ^{137}Cs -bearing soil particles that reached Japan in March 2002 came from somewhere in Mongolia or northeastern China, where there are no known nuclear test sites or nuclear facilities. The source area for the dust associated with the March 2002 sandstorms is typically covered by grassland and shrubs, and only a small part of the area is desert (Fig. 8).

(3) Radioactivity of grassland soils in northeastern China

We collected soil samples for radioactivity measurement (Fig. 3) from within an area in northeastern China where dust-raising events were frequently observed in March 2002 (Fig. 8).

^{137}Cs inventories were in the range of 176.0–3710 Bq m⁻² (Table 2) and showed a positive correlation with mean annual precipitation (Fig. 9), which suggests that the ^{137}Cs originated as global fallout from past atmospheric nuclear testing. Global fallout studies have shown that around 90% of the total deposition of ^{137}Cs occurs as wet deposition (UNSCEAR, 2000a). High rainfall during the passage of a radioactive cloud generated by a nuclear accident also results in much deposition of anthropogenic radionuclides (Brenk and Vogt, 1981; UNSCEAR, 2000b). However, since most such deposition usually takes place within a few days of the accident, its correlation with annual precipitation is weak. The distribution of deposited radionuclides due to local fallout relates to the track of radioactive clouds generated by a nuclear accident or by nuclear testing (UNSCEAR, 2000b; Simon et al., 2006). Thus, the accumulated ^{137}Cs in the samples of our study apparently did not originate from past local fallout.

Deposited ^{137}Cs is strongly adsorbed onto surface soil particles, especially clay and organic matter, and its movement

Table 2. ^{137}Cs inventory, mean annual precipitation, and soil characteristics of sampling sites in northeastern China.

Site	^{137}Cs inventory (Bq m^{-2})	Mean annual precipitation (mm)	Organic matter (%)			Clay+Silt (%)			Clay (%)			Silt (%)		
			0-2 cm	2-4 cm	4-6 cm	0-2 cm	2-4 cm	4-6 cm	0-2 cm	2-4 cm	4-6 cm	0-2 cm	2-4 cm	4-6 cm
S1	176.0	239	1.1	3.1	3.3	9.0	22.0	22.3	5.6	13.5	13.4	3.4	8.5	8.9
S2	308.7	199	1.1	1.4	1.6	14.2	13.0	13.6	10.9	10.1	10.6	3.3	2.9	3.0
S3	832.5	181	1.2	0.8	1.1	5.6	8.5	11.6	3.5	6.0	8.3	2.1	2.4	3.2
S4	1210	202	1.7	3.0	1.0	13.0	22.7	23.9	8.1	14.4	15.4	4.9	8.4	8.6
S5	1156	250	1.6	2.3	2.2	14.5	22.1	19.2	10.3	15.0	13.4	4.1	7.1	5.8
X1	3199	251	3.4	4.9	4.6	12.9	22.9	26.1	8.5	12.9	15.5	4.4	10.0	10.6
X2	3657	262	4.1	3.7	2.2	12.7	16.0	12.6	8.6	10.1	8.9	4.1	5.8	3.7
X3	1777	273	3.2	3.1	2.5	19.6	16.3	16.5	12.9	11.2	11.6	6.8	5.2	4.9
X4	1539	282	10.5	6.1	5.5	30.2	25.0	25.1	18.4	16.0	16.4	11.8	9.0	8.7
X5	3556	306	8.9	7.8	7.0	36.5	33.7	32.1	20.3	20.6	19.9	16.2	13.1	12.2
Z1	3707	309	7.3	6.7	6.4	36.9	34.9	32.6	22.9	22.2	21.5	14.0	12.7	11.1
Z2	2907	334	5.4	4.2	4.3	24.2	24.4	22.7	14.8	15.3	14.4	9.3	9.1	8.4

Site number prefixes S, X, and Z indicate samples from Sonid-you-qi, Xilinhot, and Zhengxianbai-qi, respectively (Fig. 3). Precipitation at sampling locations was interpolated by the minimum curvature method from sites of known precipitation based on data obtained from the JMA.

depends on physical processes such as the infiltration rate of soil water and soil erosion (Ritchie and McHenry, 1990; Walling and Quine, 1991; He and Walling, 1996; Walling 1998; Li et al., 2004). Baeza et al. (1995) reported that the specific activity of ^{137}Cs increases in soil as the particle size decreases, as a consequence of the greater adsorption capacity of fine particles.

The clay and organic matter contents in the surface layer (0-2 cm) of the soil samples studied (Table 2) also showed a positive correlation with mean annual precipitation (Fig. 9). In semiarid grasslands, soil erosion caused by strong winds is the primary process of desertification, and leads to a loss of clay and organic matter in particular. Furthermore, the intensity of wind erosion is controlled by the density of the vegetation coverage (Li et al., 2005), which appears to be related to precipitation. Thus, in the past, precipitation activity not only increased ^{137}Cs fallout but also indirectly reduced the loss of soil particles and organic matter, as well as ^{137}Cs .

In our study, activities of ^{137}Cs in the surface layer (0-2 cm) were in the range of 5.5-86 mBq g^{-1} (Fig. 10) and showed positive correlations with mean annual precipitation and clay and organic matter contents (Fig. 9). These results suggest that the horizontal distribution of ^{137}Cs -bearing soil particles in grassland areas is controlled by precipitation. Moreover, these radioactivity levels are equal to or higher than the levels of 5.01-31.5 mBq g^{-1} reported by Igarashi et al. (2005) in desert soils of western China. In our study, the activities at sampling sites X3 to X5, Z1, and Z2, all in grassland areas (Fig. 3), were much higher, ranging from 46 to 86 mBq g^{-1} , than those in the desert soils. At these sites, ^{137}Cs was high in the surface layer and its activity decreased exponentially with depth. Clay and silt contents at sampling sites X3 to X5, Z1, and Z2 (Table 2) were also high in the surface layer (0-2 cm), and equal to or higher than those in deeper subsurface soil at the same sites. Thus, we inferred that the loss of soil particles from the surface by wind erosion was negligible at these sites.

However, at sampling sites X1 and X2, ^{137}Cs concentrations in the surface layer were lower than those in the deeper subsurface soil (Fig. 10). This distribution can be explained by soil erosion or sedimentation at these sites. Clay and silt contents in the surface layer were also lower than those in the deeper subsurface soil at sample sites X1 and X2. As ^{137}Cs inventories at these two sites were high (3199 and 3657 Bq m^{-2}), the loss of ^{137}Cs -bearing soil particles was apparently negligible. Therefore, these vertical distributions of ^{137}Cs and mineral particles can be most easily explained by the deposition of aeolian sand in the area of sites X1 and X2 sometime after the nuclear testing period. Sites X1 and X2 are in western Xilinhot, and sand may have been transported by strong winds from farther west, where we assume that wind erosion is active. Although clay and fine silt suspended in the air can be carried by wind for great distances before they are deposited, aeolian sand tends to be deposited close to and downwind from their source (Liu et al., 2005). ^{137}Cs

concentrations near the surface may thus be diluted by the addition of sand to the surface layer.

At sites S1 to S5, ¹³⁷Cs activities in the soil surface layer and ¹³⁷Cs inventories were in the ranges of 5.5-21 mBq g⁻¹ and 176.0-1210 Bq m⁻², respectively, and were lower than at the other sampling sites. At these sites, the smaller amounts of ¹³⁷Cs deposited following past nuclear tests can be attributed to the lower precipitation in this area. Loss of ¹³⁷Cs-bearing soil particles by wind erosion might also explain the low ¹³⁷Cs concentrations at these sites. Clay and silt contents in the surface layer were lower at sites S1 to S5 than at sites X3 to X5, Z1, and Z2, where wind erosion was negligible. Sites S1 to S5 are near the Gobi, so it is likely that wind erosion and the resultant desertification have had considerable effect in this

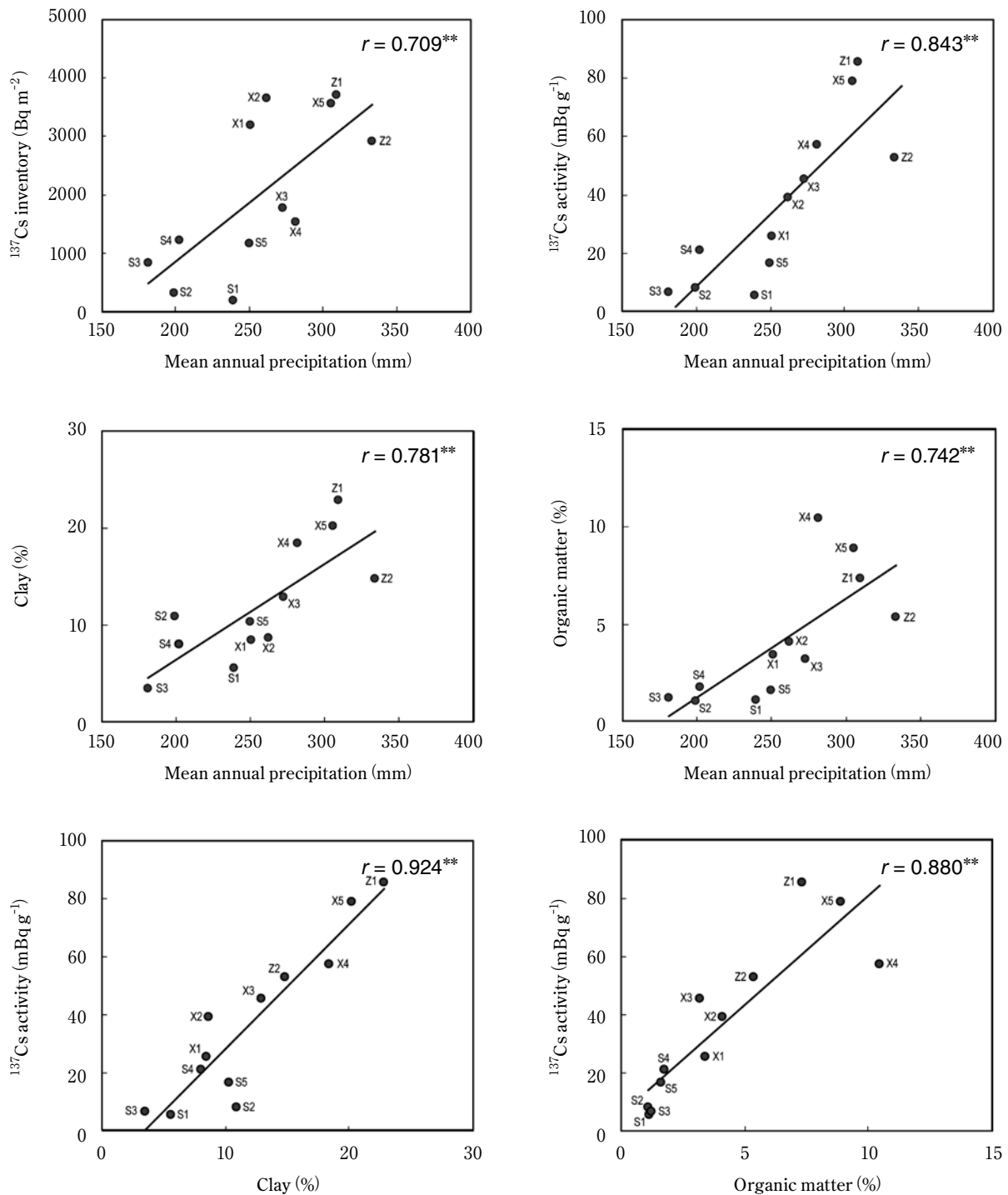


Fig. 9. Relationships among ¹³⁷Cs inventory (area-based total activity), mean annual precipitation, and contents of clay, organic matter and ¹³⁷Cs in the surface layer (0-2 cm) of the sampled soils.

area.

The distribution of ^{137}Cs in grassland soils is strongly related to the intensity of wind erosion, which is related to the vegetation cover (Zhang et al., 2002; Hu et al., 2005). Thus, ^{137}Cs -bearing soil particles are likely to be removed from eroding grasslands where vegetation coverage is weak, and suspended in the atmosphere. In contrast, ^{137}Cs concentrations are preserved in the surface soil of stable grasslands. However, a rapid and severe reduction of vegetation cover in previously stable grasslands would lead to a massive release of accumulated ^{137}Cs . Therefore, stable grasslands, if the vegetation cover becomes reduced, are potential sources of ^{137}Cs -bearing soil particles.

Kurosaki and Mikami (2003) have suggested that frequent strong winds have been the primary cause of major Asian dust events in recent years. They also reported that snow cover increases the threshold wind velocity of dust suspension (Kurosaki and Mikami, 2004). However, because sand and dust storms are not only weather phenomena but also geomorphic processes (Xu, 2006), an increase in the frequency of dust events may be caused by a reduction in vegetation cover in the grassland area. Denser vegetation cover in summer tends to reduce the frequency of sandstorms in the following spring (Zou and Zhai, 2004). Furthermore, Liu et al. (2004) suggested that more precipitation in the preceding

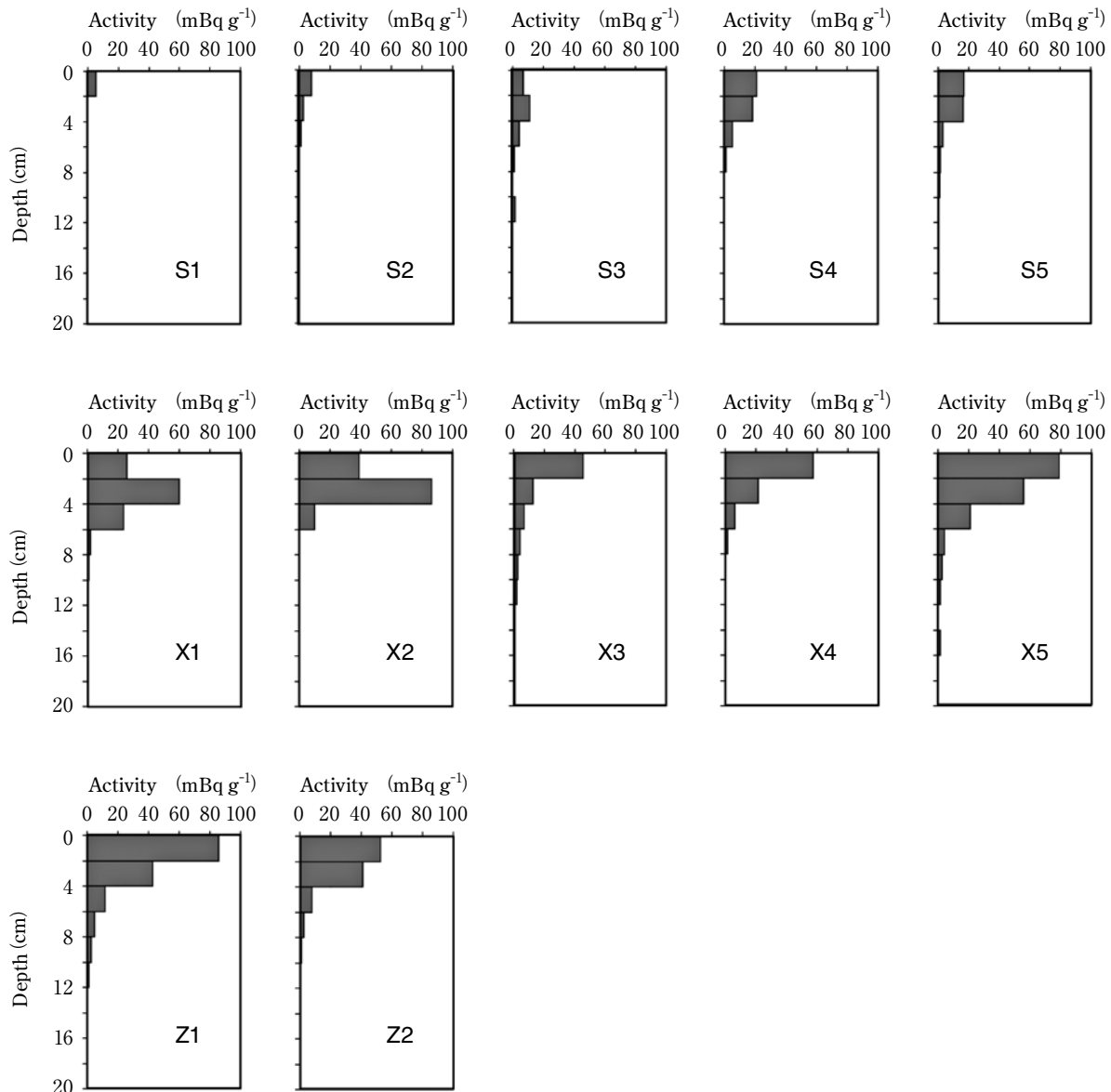


Fig. 10. Vertical distribution of ^{137}Cs activity in grassland soils at sampling sites in northeastern China.

year leads to more flourishing vegetation and lower dust emission in the following spring, especially in areas close to the China-Mongolia border.

Most of northern China has experienced prolonged dry periods since the late 1990s (Zou et al., 2005). Zou et al. (2005) determined that northeastern China experienced its most extensive droughts in 2001 and 2002; nearly 70% of the region experienced drought conditions in these years. These dry conditions reduced the vegetation cover of the grasslands. The frequency of sandstorms in northeastern China in recent years and the accompanying prodigious release of ^{137}Cs -bearing soil particles from grassland soils into the atmosphere are attributable to this change.

4. Conclusions

Monthly records of ^{137}Cs deposition at Aomori, Japan, during the 1990s show a consistent seasonal variation, with higher levels of deposition occurring in spring. In March 2002, unexpectedly high deposition associated with an Asian dust event was observed over the northwestern coastal area of Japan. Analysis of land-based weather data showed that sandstorms and other dust-raising phenomena occurred in March 2002 in areas of Mongolia and northeastern China characterized by grasslands and shrubs. Moreover, radioactivity measurements showed ^{137}Cs enrichment in the surface layer of grassland soils in northeastern China, which we believe accumulated during past atmospheric nuclear testing. These results suggest that these grassland areas are a potential source of ^{137}Cs -bearing Asian dust. Most of northern China has experienced prolonged dry periods since the late 1990s. In 2001 and 2002, there were severe droughts in northeastern China. The massive sandstorms of recent years and the accompanying prodigious release of ^{137}Cs -bearing soil particles were the result of erosion by seasonal strong winds of grasslands whose vegetation cover had been greatly reduced by drought.

Chapter III

Relative contributions of Asian dust and local dust to atmospheric deposition of ^{137}Cs in Japan

1. Introduction

In the previous chapter, we showed that recent ^{137}Cs deposition observed in Japan is attributable to soil dust transported from the East Asian continent. However, soil dust is also raised around the Japanese monitoring sites by the strong winds that are common in spring, and this locally raised dust might also contribute to the deposition of ^{137}Cs (Igarashi et al., 2001). The relative contributions of Asian dust and local dust to the atmospheric deposition of ^{137}Cs have been estimated previously, mostly on the basis of monthly monitoring. Because dust events are short-term phenomena of a few days' duration (Zhou and Zhang, 2003; Iino et al., 2004), it has been difficult to determine the contributions of individual dust events by conventional monitoring. Therefore, to evaluate radionuclide deposition in Japan with more precision, it is essential to observe atmospheric deposition with a temporal resolution that is high enough to resolve the effects of Asian and local dust events. Our objective in this chapter is to estimate the relative contribution of these dust phenomena to the total ^{137}Cs deposition. For this purpose, we observed weekly atmospheric deposition in spring.

2. Data and methods

(1) Collection of atmospheric deposition samples

We collected atmospheric deposition samples weekly for eight weeks from March to April 2007 at the National Institute for Agro-Environmental Sciences (NIAES) in Tsukuba, Japan (Fig. 11). Tsukuba is on the Pacific Ocean side

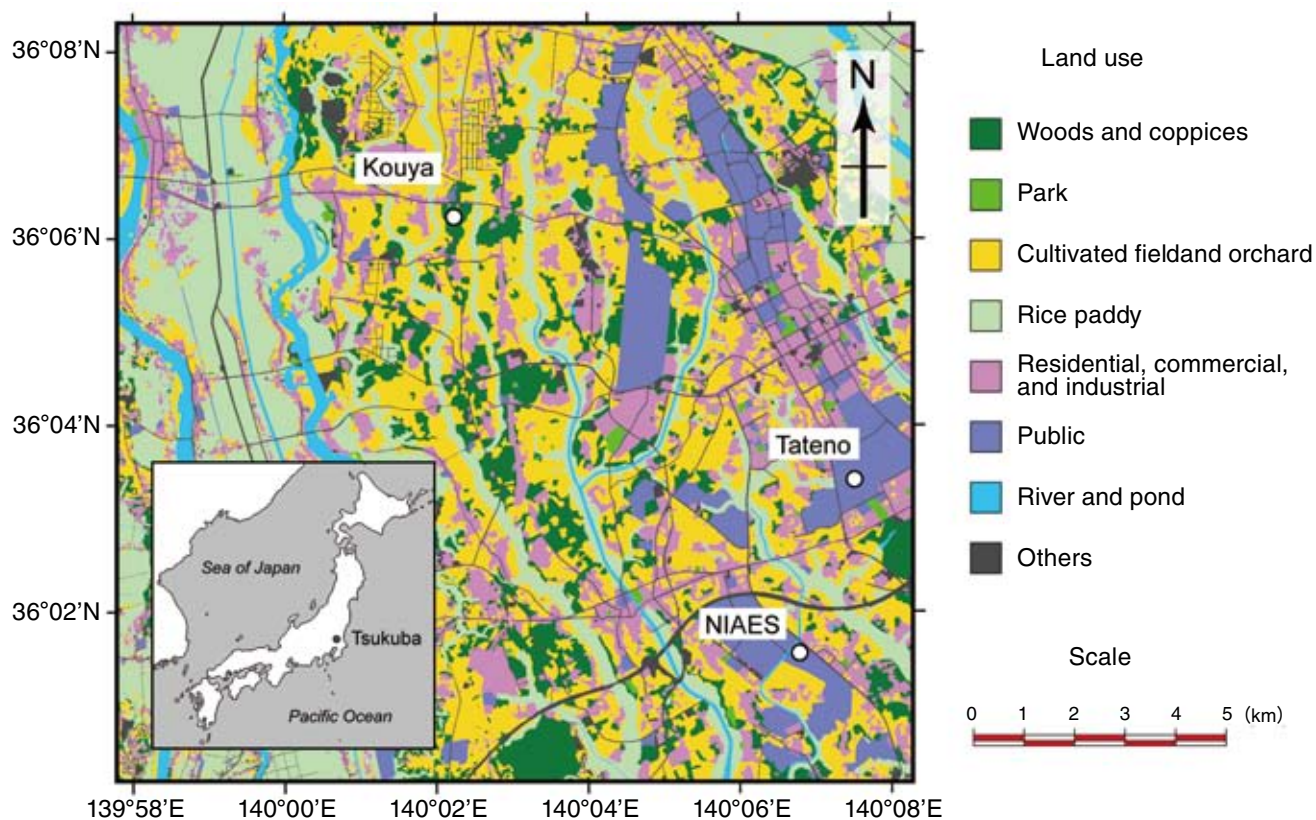


Fig. 11. Sampling sites for atmospheric deposition at NIAES, the SPM monitoring station at Tsukuba-Kouya, and the meteorological station at Tsukuba-Tateno. Colors represent land-use types, which are derived from Detailed Digital Land Use Information (Geographical Survey Institute, 1998).

of the Japanese Islands and is a typical Japanese suburban site that is surrounded by rice paddies, cultivated fields, residential areas, and small woods and coppices. Six stainless-steel open-surface collectors with a total surface area of 1.18 m^2 were installed 1 m above ground level on the observation field. The collectors were filled to the depth of 1 cm with distilled water to prevent re-suspension of deposits. The samples, which resulted from both wet and dry deposition, were transported to the laboratory at NIAES and evaporated to dryness at 80°C in evaporation dishes placed on hot plates.

(2) Analysis of deposition samples

After being dried in an oven at 110°C , the deposition samples were weighed. Their radioactivities were determined by using gamma spectrometry with a well-type high-purity germanium detector (Canberra GCW) with an efficiency of 26% and a multi-channel analyzer. The gamma emission peaks for ^{137}Cs at 661 keV were used for these measurements. The count time was typically about 160 ks. The detection limit was defined as 3σ of the counted value (Cooper, 1971). The ^{137}Cs activities were corrected for the decay that took place between the date of sample collection and the sample analysis. Total deposition of ^{137}Cs , expressed in units of mBq m^{-2} , was calculated by dividing the ^{137}Cs activity by the surface area of the collector. In addition, the mineral contents of the samples were determined after removing soluble salts by dissolution in 1 M HCl and then ashing at 800°C to remove organic material (Nelson and Sommers, 1996). ^{137}Cs specific activity was defined as the ratio of ^{137}Cs activity to the mineral content and expressed in units of mBq g^{-1} .

(3) Monitoring data of SPM and surface meteorological factors

SPM concentration data collected at Tsukuba-Kouya station, which is 10 km from NIAES, were provided by the Environmental Management Division, Department of Civil Affairs and the Environment, Ibaraki Prefectural Government (Fig. 11).

We obtained nationwide observation data of Asian dust events from the JMA web site (Japan Meteorological Agency, 2008). These data are based on visual observations at 85 weather stations in Japan. We defined the frequency of observed Asian dust events as the total number of weather stations that observed aeolian dust or dust haze on a given day. We also obtained hourly surface meteorological data collected at the Tsukuba-Tateno station (Fig. 11), including precipitation and wind speed data, from the JMA web site. We defined the strong-wind frequency as the number of observations of wind speed greater than 5 m s^{-1} in an observation week as a percentage of the total number of wind speed observations in that week. Furthermore, we determined and mapped the spatial distribution of dust-reporting sites in Japan from surface meteorological data. We obtained six-hourly (four times a day) meteorological data based on SYNOP (surface synoptic observations) reports for East Asia, available on CD-ROM (Japan Meteorological Business Support Center, 2007) from the JMA. (SYNOP is a numerical code used for reporting weather observations, and SYNOP reports are typically sent worldwide every three hours from land-based weather stations.) The code number 06 in current SYNOP reports indicates observations of dust suspended in the air. However, the records from Tsukuba were insufficient for our use.

3. Results and discussion

(1) Variations in the deposition of mineral particles and SPM concentrations

During the first five weeks of observation at the Tsukuba monitoring site, $0.67\text{--}1.48 \text{ g m}^{-2}$ of mineral particles was deposited per week (Fig. 12). These relatively large amounts suggest that during this period either dust concentrations in the atmosphere were high or atmospheric conditions were particularly conducive to deposition.

In the fifth week of observation, from 30 March to 6 April 2007, 0.76 g m^{-2} of particles was deposited (Fig. 12). During this week, high SPM concentrations of up to $107 \mu\text{g m}^{-3}$ were observed from 1 to 3 April (Fig. 13). In addition, relatively high SPM concentrations of up to $79 \mu\text{g m}^{-3}$ were observed during the fourth week, from 23 to 30 March, when 1.22 g m^{-2} of particles was deposited. The samples deposited during the fourth and fifth weeks, when an increase in SPM was also observed, possibly consisted of Asian dust. It is well known that the SPM concentrations in Japan reflect dust transported

from the East Asian continent, and that increases in SPM concentrations observed during environmental air monitoring at weather stations reflect contributions of Asian dust (Zhang et al., 2006). Because larger particles fall quickly out of the atmosphere under the influence of gravity, the dust that is carried over long distances in the upper atmosphere to Japan generally consists of smaller particles with a diameter of less than $10\ \mu\text{m}$ (Mori et al., 2003).

During the first three weeks of our observations, from 2 to 23 March, substantial deposition of mineral particles, $0.67\text{--}1.48\ \text{g m}^{-2}$ per week, was observed (Fig. 12), although there was no notable increase in SPM (Fig. 13). The infrequent precipitation during these weeks (Fig. 13) suggests that atmospheric conditions were not conducive to wet deposition of suspended particles. In these weeks, measurable precipitation of $2.0\text{--}12.0\ \text{mm}$ was recorded on just one day in each week. Since SPM observations and atmospheric conditions do not support enhanced wet deposition, dry deposition from locally raised soil dust is the only plausible explanation for the observed mineral deposition during these three weeks. Wind erosion of soil can cause soil particles of up to $100\ \mu\text{m}$ in diameter to become suspended in the air (Hudson, 1986), and soil dust raised around a monitoring site by strong winds would likely be dominantly coarse ($>10\ \mu\text{m}$ in diameter). Therefore, coarse local dust particles would be represented in the total mineral particle content of the deposition samples.

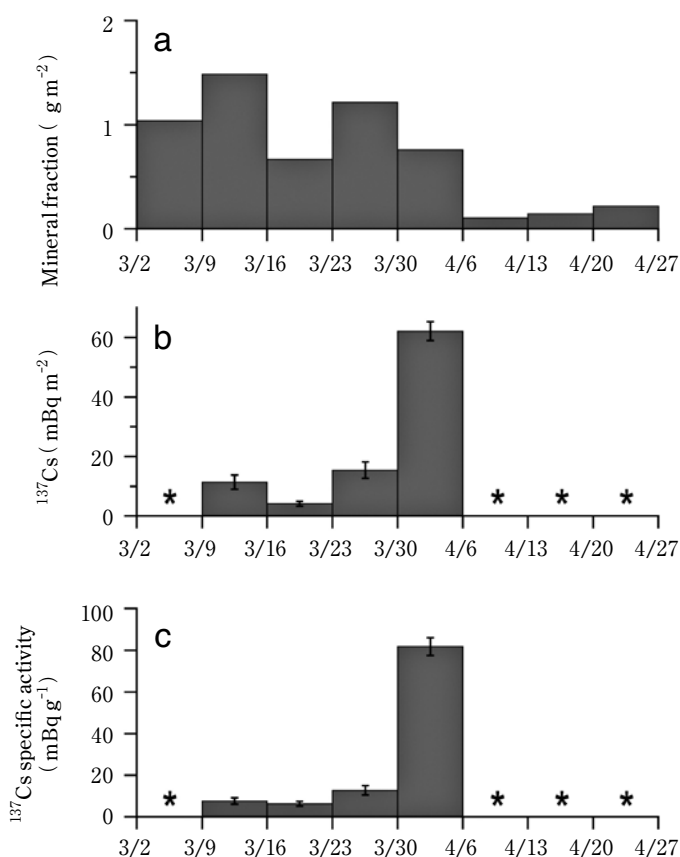


Fig. 12. Weekly atmospheric deposition of mineral particles (a) and ^{137}Cs (b), and weekly ^{137}Cs specific activity measurements (c) at Tsukuba from 2 March to 27 April 2007. Deposits were collected once a week for eight weeks. Notable ^{137}Cs activity (calculated per unit weight of mineral particles in the total deposit) was detected four times over the monitoring period. Asterisks indicate weeks when no ^{137}Cs was detected. Error bars show counting error.

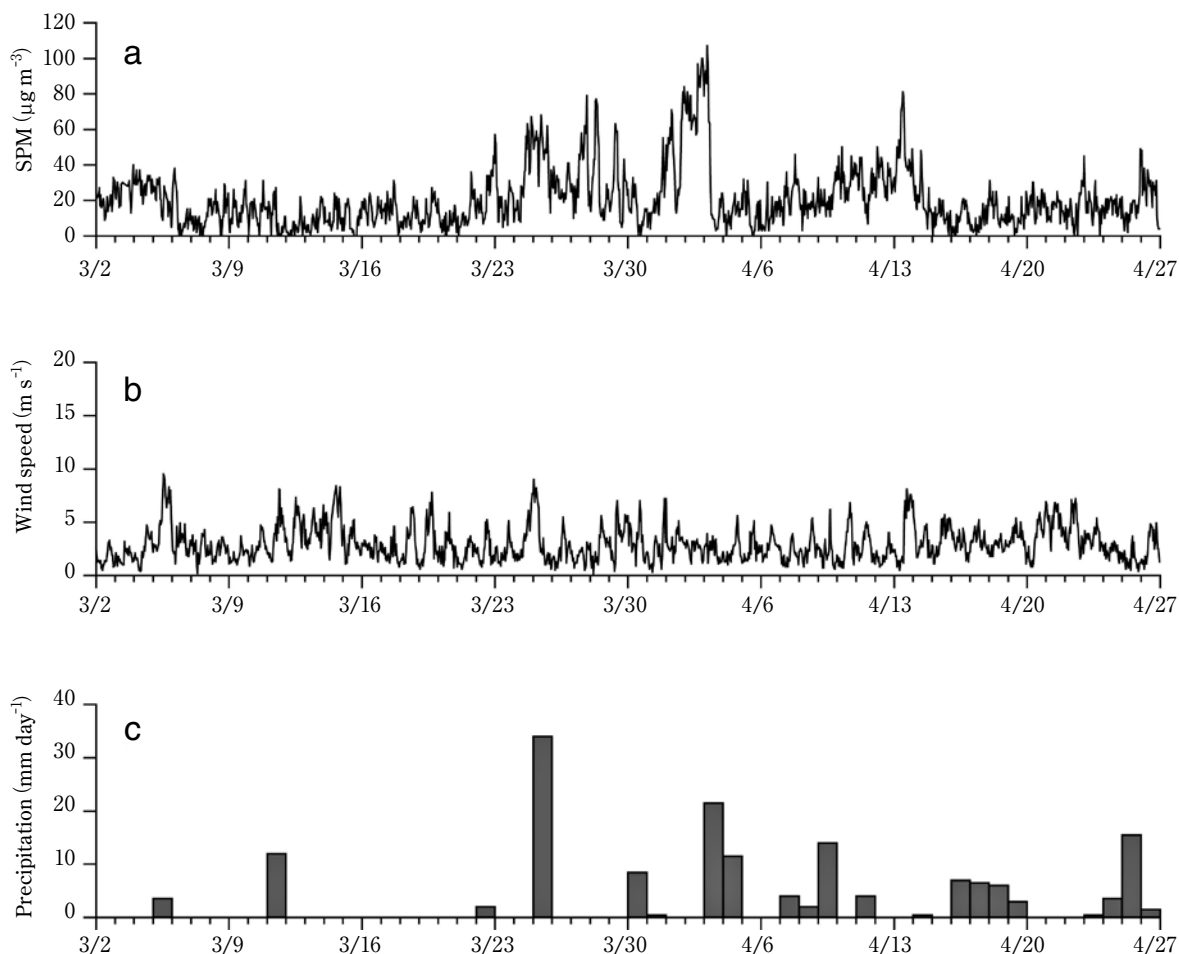


Fig. 13. SPM concentrations and meteorological information related to atmospheric deposition from 2 March to 27 April 2007. Hourly SPM concentrations (a) were observed at Tsukuba-Kouya. Hourly wind speed (b) and daily precipitation (c) were observed at Tsukuba-Tateno. High SPM concentrations of up to $107 \mu\text{g m}^{-3}$ were observed continuously from 1 to 3 April in the fifth week.

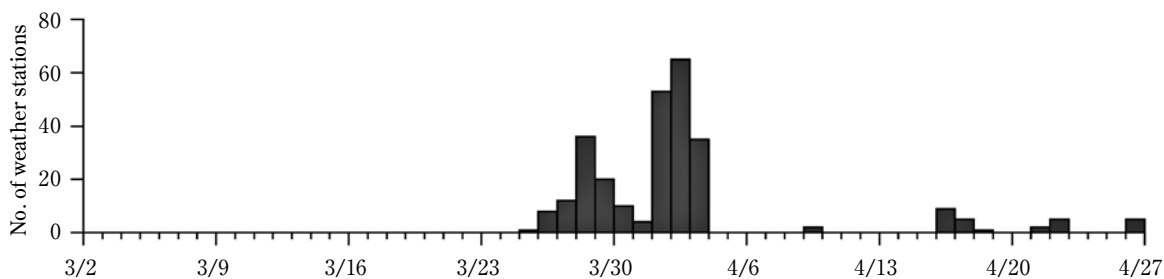


Fig. 14. Asian dust observations in Japan during the monitoring period from 2 March to 27 April 2007. The frequency of Asian dust observations was defined as the number of Japanese weather stations that observed aeolian dust or dust haze on a particular day, as determined by ground-based visual observations. Two clear dust peaks occurred during this period, one on 28 March and the other on 2 April.

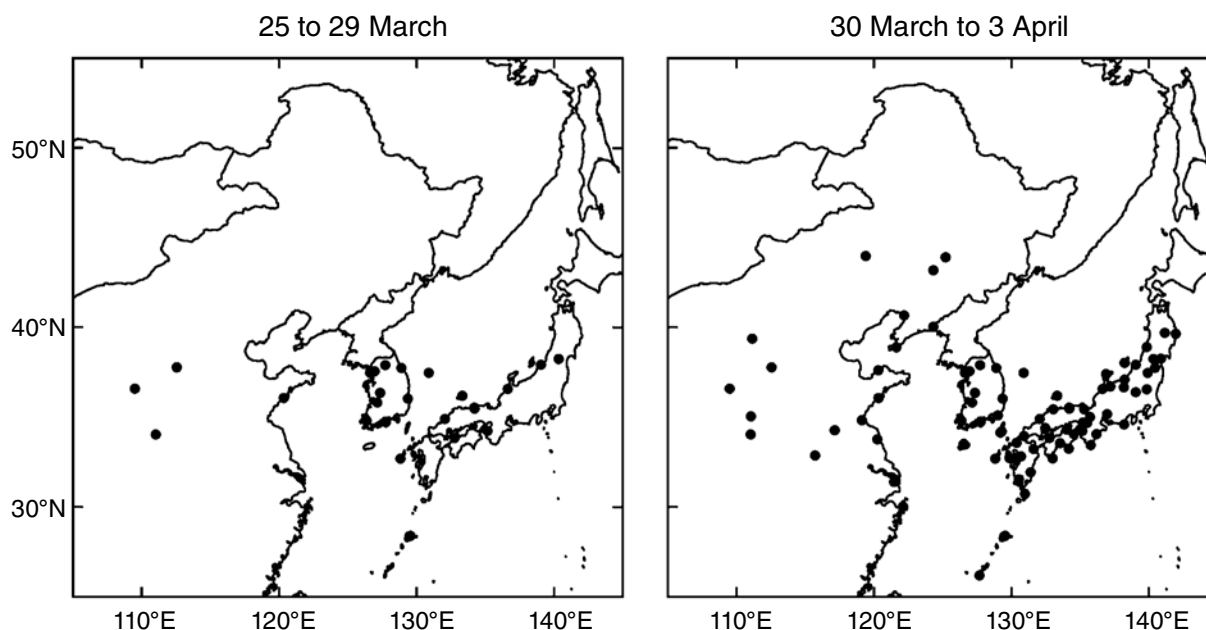


Fig. 15. Spatial distribution of dust-reporting sites in spring 2007, based on meteorological data contained in SYNOP (surface synoptic observation) reports.

Table 3. Summary of surface meteorological conditions during the monitoring period.

Period	Date of sampling	Precipitation (mm)	Frequency of strong wind* (%)	Maximum wind speed (m s^{-1})
1	3/9	3.5	8.3	9.5
2	3/16	12.0	23.2	8.4
3	3/23	2.0	8.9	7.8
4	3/30	42.5	15.5	9.0
5	4/6	33.5	5.4	7.2
6	4/13	24.0	5.4	6.8
7	4/20	23.0	13.1	5.7
8	4/27	21.0	17.9	7.2

* Expressed as the number of observations of an hourly wind speed greater than 5 m s^{-1} in an observation week as a percentage of the total number of wind speed observations in that week. Hourly wind speed was defined as the average over the ten minutes before the observation time.

(2) Meteorological factors related to atmospheric deposition

Information on aeolian dust from ground-based visual observations includes some uncertainty, but such observations are nevertheless useful for detecting Asian dust events (Iino et al., 2004). Asian dust was observed in Japan continuously from 25 March to 3 April 2007 (Fig. 14). Two clear dust peaks occurred during this period, one on 28 March and the other on 2 April. Because the spatial distribution of dust-reporting sites in Japan from 30 March to 3 April (Fig. 15) indicates that suspended dust was observed around Tsukuba, the marked increase in the SPM concentration from 1 to 3 April (Fig. 13) and the increased deposition of mineral particles observed during the fifth week (Fig. 12) can both be attributed to Asian dust. Although dust was not observed around Tsukuba from 25 to 29 March (Fig. 15) in the fourth week, dust was observed in other areas of Japan in that week, suggesting some possible contribution of Asian dust to the deposition in that week as well.

As previously mentioned, precipitation was infrequent during the first three weeks of our observations (Fig. 13), not only indicating that atmospheric conditions were not conducive to the deposition of Asian dust but also suggesting that the surface soil close to the monitoring site was likely dry and readily able to produce local soil dust. In particular, in the second week, the strong-wind frequency was 23.2% (Table 3) and the maximum deposition of 1.48 g m^{-2} was recorded, despite there being little evidence for Asian dust. Higher deposition associated with the increased frequency of strong winds suggests that local dust dominantly accounted for the deposition at that time.

Local dust likely also contributed to the deposition during the fourth week, as suggested by the discontinuous precipitation and the frequent strong winds. In this week, precipitation on a level with that during the previous three weeks was recorded on just one day (Fig. 13), and the strong-wind frequency reached 15.5% (Table 3). Thus, both Asian dust and local dust could have contributed to the deposition during the fourth week.

It can be assumed that the surface soil around the Tsukuba site was relatively wet during the fifth week, as 33.5 mm of precipitation (Table 3) fell over four days (Fig. 13). Further, the strong-wind frequency in the fifth week was only 5.4%, the lowest frequency during the entire monitoring period (Table 3). Under these conditions, little local dust would have been raised to contribute to the total deposition. From this evidence, it is reasonable to infer that the deposition during the fifth week was primarily derived from Asian dust.

During the last three weeks of our observations, from 6 to 27 April, relatively small amounts of deposition were observed ($0.11\text{--}0.22 \text{ g m}^{-2}$; Fig. 12). During this period, the wetness of the surface soils, a result of the frequent precipitation during this period (Fig. 13), would have limited the amount of local dust that was raised and subsequently re-deposited. Although high SPM concentrations of up to $81 \mu\text{g m}^{-3}$ were briefly observed on 13 April, these high levels cannot be attributed to a particular soil dust source, because no Asian dust was reported in the meteorological observations for that day.

In summary, during the monitoring period, the weekly deposition amount could be attributed to Asian dust or local dust, or both. The relative contribution of local dust increased when the local surface soil was dry, during periods of low precipitation. Conversely, when the surface soil around the site was wet, little local dust was raised even by strong winds, and deposition of local dust was therefore small.

(3) Variations in atmospheric ^{137}Cs deposition and specific activity in relation to potential sources

^{137}Cs deposition during the fifth week, when an Asian dust event was observed around Tsukuba, was 62.3 mBq m^{-2} , and accounted for 67% of the total ^{137}Cs deposition during the entire monitoring period (Fig. 12). This result suggests that most ^{137}Cs deposition in spring is derived from Asian dust events. Moreover, a high specific activity of ^{137}Cs (81.8 mBq g^{-1}) was also observed at this time. As ^{137}Cs is preferentially adsorbed onto the fine fraction (e.g., clay-sized particles) in the soil (Ritchie and McHenry, 1990), ^{137}Cs specific activity might be high in deposits of Asian dust because of the high content of fine-grained mineral particles in this dust.

Baeza et al. (1995) also reported that the specific activity of ^{137}Cs increases in soil as the particle size decreases, as a consequence of the greater adsorption capacity of fine particles. In the case of an Alumi-Haplic Acrisol in South China, for example, ^{137}Cs was detected only in the particle-size fraction of less than $50 \mu\text{m}$ (Li et al., 2004). Furthermore, Li et al. (2004) confirmed that ^{137}Cs activity in the clay fraction ($<2 \mu\text{m}$) and the very fine silt fraction ($2\text{--}5 \mu\text{m}$) was always much higher than that in other fractions. During transport of dust from its continental Asian source, the coarse fraction (sand and coarse silt) falls to the ground under the influence of gravity sooner than the smaller particles, which are more likely to reach Japan. Mori et al. (2003) investigated the changes in the particle-size distribution of Asian dust that occur during long-range transport. They showed that the size distribution of dust samples collected in Beijing, close to the dust source, had a major peak at $4.7\text{--}7.0 \mu\text{m}$, in the coarse particle-size range, whereas that of aerosols collected at Yamaguchi in western Japan during the same dust event had two peaks, one at $0.43\text{--}0.65 \mu\text{m}$ and another at $3.3\text{--}4.7 \mu\text{m}$. Thus, the higher ^{137}Cs specific activity observed in the deposits of Asian dust can be explained by enrichment due to particle-size

sorting during transport.

In contrast, during the first three weeks of our investigation, when local soil dust dominantly accounted for the deposition, the weekly ^{137}Cs deposition was less than 11.4 mBq m^{-2} , demonstrating that local dust contributed less to ^{137}Cs deposition than did Asian dust events (Fig. 12). ^{137}Cs specific activities were also low, less than 7.7 mBq g^{-1} , during the first three weeks of observation. These relatively low activities are attributable to both the dependence of ^{137}Cs specific activity on particle size and the ^{137}Cs activity in the source soils, as with the deposits derived from Asian dust. The relative coarseness of local dust particles near the monitoring site implies that local dust contains fewer ^{137}Cs -bearing soil particles than Asian dust. Cultivated fields that are bare in spring are potential sources of local dust (Igarashi et al., 2001). The ^{137}Cs activity of the deposits during the first three weeks of observation was comparable to that of surface soil in a cultivated field near the Tsukuba site, where the recently measured ^{137}Cs activity in the soil ranges from 6.3 to 7.5 mBq g^{-1} (Komamura et al., 2005). Thus, low activity in the source soil can also explain the low ^{137}Cs activity in the deposits during these weeks.

Although both Asian dust and local dust may have contributed to the deposition during the fourth week, ^{137}Cs specific activity during that week remained at 12.6 mBq g^{-1} , close to the level of local dust deposits (Fig. 12). This low activity is attributable to a relatively small contribution of Asian dust.

These results suggest that recent ^{137}Cs deposition in Japan is attributable to both transport of dust raised by Asian dust events and dry deposition of locally derived dust. However, the contribution of local dust was considerably smaller than that of Asian dust, indicating that the primary source of atmospheric ^{137}Cs in Japan is dust transported from the East Asian continent.

4. Conclusions

Weekly atmospheric deposition of mineral particles increased both when SPM levels were high and when local conditions were dry and windy. Thus, such deposition is attributable to both transport of dust raised by Asian dust events and dry deposition of locally derived dust. During the week when an Asian dust event was observed, atmospheric ^{137}Cs deposition reached 62.3 Bq m^{-2} and accounted for 67% of the total deposition of ^{137}Cs during the entire monitoring period. Thus, the primary source of ^{137}Cs deposition in spring is likely to be dust transported from the East Asian continent. During the same week, ^{137}Cs specific activity reached 81.8 mBq g^{-1} . On the other hand, the specific activity of ^{137}Cs in locally derived dust deposits was much lower, and local dust contributed little to the total deposition of ^{137}Cs over the monitoring period. The higher activity of deposits derived from Asian dust may be due to enrichment as a result of grain-size sorting during transport.

Chapter IV

General features of atmospheric deposition of ^{137}Cs -bearing Asian dust

1. Source area of ^{137}Cs -bearing Asian dust

The primary objective of this study was to clarify the source of ^{137}Cs deposited in Japan in relation to continental Asian dust phenomena. In chapter I, our literature review and observation data of ^{137}Cs deposition in Japan suggested that most ^{137}Cs has a continental source. Furthermore, in chapter II, we used surface meteorological data and radioactivity measurements of continental soils to determine the specific source areas of ^{137}Cs -bearing Asian dust.

The desert areas of western China have been regarded as the primary source of Asian dust (Xuan and Sokolik, 2002), and in the early 2000s, mechanisms of sand and dust suspension in those areas, dust transport processes, and the influence of the dust on global climate were investigated by several research programs, including the Asian Pacific Regional Aerosol Characterization Experiment (ACE-Asia) and the Transport and Chemical Evolution over the Pacific (TRACE-P) programs. In these programs, a number of ground-based observation sites with advanced observation equipment such as aerosol lidars were established in the arid areas of western and northern China, Korea, and Japan (Iwasaka et al., 2003; Zhang et al., 2003; Huebert et al., 2003). As 22 atmospheric nuclear tests were conducted at Lop Nor (40°N , 90°E), in the desert of western China, between 1964 and 1980 (UNSCEAR, 2000a), Igarashi et al. (1996) suggested that the source of recent ^{137}Cs deposition in Japan was in that area.

However, in the early 2000s, Asian dust events became unusually frequent in the eastern part of the Asian continent, and they were less frequent in the deserts of western China (Shao and Wang, 2003; Kurosaki and Mikami, 2003). Over the last 40 years in China, dust event frequencies in general have been decreasing (Parungo et al., 1994; Sun et al., 2000; Qian et al., 2002; Yoshino, 2002), but severe dust storms were observed most frequently over Mongolia and Inner Mongolia during 2000 to 2002 (Shao and Wang, 2003). Kurosaki and Mikami (2003) confirmed that sandstorms and other dust-raising events occurred more frequently in the eastern part of the Asian continent during 2000 to 2002 than during the previous seven years.

The results described in chapter II showed that sandstorms and other dust-raising phenomena in March 2002 were observed most frequently in the semiarid grasslands of Mongolia and northeastern China, which are far from the nuclear test site at Lop Nor in the deserts of western China (Fig. 8). Furthermore, radioactivity measurements showed ^{137}Cs enrichment in the grassland soils of northeastern China (Table 2). Because ^{137}Cs inventories were positively correlated with mean annual precipitation (Fig. 9), the source of the accumulated ^{137}Cs in the soils likely was global fallout from the tropopause and stratosphere caused by past nuclear testing. This correlation also suggests that past local fallout originated from an accidental release or from Chinese nuclear testing at Lop Nor can be neglected because the correlation of annual precipitation with ^{137}Cs deposition due to local fallout is weak. The semiarid grasslands in the eastern part of the Asian continent are potential sources of ^{137}Cs -bearing soil particles. Therefore, we inferred these grasslands to be the primary source of ^{137}Cs -bearing dust in recent years. A possible explanation for the recent increase in the frequency of dust events is the reduction in vegetation cover caused by drought. However, the changes in land surface conditions in these areas and subsequent dust suspension require further investigation, because other studies have regarded arid areas in western and northern China as the primary source of Asian dust (Xuan and Sokolik, 2002; Zhang et al., 2003).

Overgrazing by the increasing numbers of livestock in Mongolia and northeastern China has led to extensive degradation of grassland in those regions (Li et al., 2005). Desertification of the East Asian continent in response to climate change and grassland degradation will likely result in an increase in ^{137}Cs -bearing soil particles in the atmosphere, and their subsequent re-deposition.

2. Relative contributions of dust phenomena to deposition of ^{137}Cs in Japan

Recent ^{137}Cs deposition observed in Japan is attributable to soil dust transported from the East Asian continent. However, Rosner et al. (1997) suggested that re-suspended anthropogenic radionuclides might also have local sources near observation sites. Soil dust is raised around the Japanese monitoring sites by the strong winds that are common in spring, and this local dust might also contribute to the observed deposition of ^{137}Cs .

The results described in chapter III showed that during a week when an Asian dust event was observed, atmospheric ^{137}Cs deposition reached 62.3 mBq m^{-2} and accounted for 67% of the total deposition of ^{137}Cs during the entire eight-week monitoring period (Fig. 12). This result indicates that the primary source of ^{137}Cs deposition in spring is likely dust transported from the East Asian continent. Moreover, a high specific activity of ^{137}Cs (81.8 mBq g^{-1}) was also observed during this week (Fig. 12). The high activity of the deposits derived from Asian dust may be due to enrichment as a result of grain-size sorting during transport. On the other hand, the specific activity of ^{137}Cs in locally derived dust deposits was much lower, indicating that local dust contributed little to the total deposition of ^{137}Cs over the monitoring period.

The primary source of the local dust is unknown and requires further investigation, although cultivated fields, which are bare in spring, are potential sources of local dust. Moreover, the contribution of local dust varies depending on the surrounding environment and local climate. Hirose and Sugimura (1984) found seasonal changes in the atmospheric concentration of Th isotopes. Igarashi et al., (2005) also found high $^{137}\text{Cs}/^{90}\text{Sr}$ activity ratios of deposition samples collected in winter and spring. These seasonal differences in radioisotopes reflect changes in their sources. In particular, the difference in $^{137}\text{Cs}/^{90}\text{Sr}$ activity ratios of soil particles may provide a clue as to the source of the dust (Igarashi et al., 2001). Additional isotope ratio measurements would be useful for ascertaining more detailed characteristics and contributions of Asian and local dust.

3. Contribution of ^{137}Cs -bearing Asian dust to the atmospheric environment

We determined grasslands in the eastern part of the Asian continent to be the primary source of ^{137}Cs deposited in Japan. The activity levels of ^{137}Cs in the surface soil of the grassland area were in the range $5.5\text{--}86 \text{ mBq g}^{-1}$ (Fig. 10), values equal to or higher than the levels in Japanese cropland soils. For example, recent ^{137}Cs activity in surface soil ranges from 6.3 to 7.5 mBq g^{-1} at Tsukuba (Komamura et al., 2005). On the other hand, these activity levels are considerably lower than the levels in contaminated soils near nuclear test sites or the Chernobyl nuclear reactor, and they seem to have little effect on human health. In 2000, the total estimated ^{137}Cs inventory in the upper 30 cm of soil within 30 km of Chernobyl in Ukraine was $2.8 \times 10^{15} \text{ Bq}$ (Kashparov et al., 2003). However, the influence of ^{137}Cs -bearing Asian dust on the atmospheric environment appears not to be negligible because dust phenomena are widespread over the East Asian continent and introduce a massive amount of soil particles into the troposphere. In fact, considerable atmospheric deposition of ^{137}Cs associated with an Asian dust event was observed over the northwestern coastal area of Japan in March 2002 (chapter II). Because Asian dust affects thickly populated areas in East Asia, investigations of ^{137}Cs transport and deposition are essential to predict future atmospheric levels of ^{137}Cs and to evaluate its effects on human health. Although our results contribute much to this purpose, further study is required for quantitative evaluation. To estimate quantitatively the effects of ^{137}Cs -bearing Asian dust, numerical modeling and simulation methods such as the Regional Atmospheric Modeling System (RAMS) and the Chemical Weather Forecast System (CFORS) would be useful (Pielke et al., 1992; Uno et al., 2003). In particular, RAMS/HYPACT and RAMS/CFORS are suitable for modeling the long-range atmospheric transport and deposition of aerosols and chemical pollutants (Uno et al., 2004; Satake et al., 2004).

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東アジアにおけるダストイベントにともなう 放射性セシウム (^{137}Cs) の大気降下

藤原英司

摘 要

^{137}Cs は核分裂反応により生成する半減期約30年の人工放射性核種で、その外部放射線による公衆被曝が放射線リスク管理上の問題となる。1950年代から70年代にかけて実施された大気圏内核実験や1986年のチェルノブイリ発電所事故によって、この ^{137}Cs が大気圏に拡散し汚染は地球全体に及んだ。その後、大気圏内核実験の実施例はなく大規模事故も発生しなかったため大気中 ^{137}Cs は減少傾向を示し、1990年代には人体への影響が想定されるような水準ではなくなった。しかし日本においては現在も ^{137}Cs の大気降下が継続し、特に春季には降下量の増大が認められる。この現象は黄砂飛来に起因すると考えられたため、 ^{137}Cs を含む砂塵の供給源や供給プロセスの解明を本研究の目的とした。

2002年3月に、北日本や日本海側地域を中心としてチェルノブイリ発電所事故時以後で最大となる顕著な ^{137}Cs 降下が観測された。2000年代前半に核爆発や原子力関連施設における重大事故等は記録されていないため、 ^{137}Cs を含む砂塵の飛来が、この現象についての唯一可能な説明であると考えられた。そこで本研究では、この事例を取り上げて研究対象とした。まず、地上実況気象通報式 (SYNOP) により報じられた天気観測データから、2002年3月における東アジア大陸部での砂塵発生事象の発生頻度分布を計算した。その結果、砂嵐等が8%以上の高頻度で観測された地点は、中国北部からモンゴルにかけての草原域に局在していることが明らかになった。このことは、同地域が2002年3月における砂塵発生の中心であったことを示す。次に、砂塵の発生が顕著であったとみられる中国内モンゴル自治区中央部の草原で採取された土壌の放射能測定を行い、 ^{137}Cs 濃度および土壌中蓄積量を求めた。その結果、草原表土における ^{137}Cs の集積が認められ、濃度は $5.5\text{--}86\text{ mBq g}^{-1}$ と、中国の核実験場に近いタクラマカン砂漠の土壌の例 ($5.01\text{--}31.5\text{ mBq g}^{-1}$) や日本の畑地土壌の例 ($6.3\text{--}7.5\text{ mBq g}^{-1}$) よりも高い水準であった。このことから、大陸の草原が ^{137}Cs を含む砂塵の供給源であると特定された。また草原土壌への ^{137}Cs 蓄積量は $176\text{--}3710\text{ Bq m}^{-2}$ であり、平均年間降水量との正の相関が認められた ($r=0.709$ 、有意水準1%)。このことは、土壌蓄積 ^{137}Cs が特定の場所からの局地的フォールアウトに由来するのではなく、グローバルフォールアウトによる累積的降下によることを強く示唆した。

土壌中 ^{137}Cs の分布は風による土壌侵食 (風食) 作用による影響を強く受け、また風食の程度は植生による土地の被覆状態と密接に関係している。つまり、植生による被覆状態が悪く風食を受けやすい土壌においては、 ^{137}Cs を含む細粒質が失われ大気へ移行しやすいのに対し、被覆状態が良く安定した草原土壌では、風食の程度は小さく表土の ^{137}Cs 濃度は高い水準で維持される。問題は、それまで安定が保たれていた草原において、何らかの理由による急激かつ深刻な植生衰退に伴い、 ^{137}Cs を含む土壌粒子の多量放出が起こる場合である。2001年から2002年にかけて中国北部は深刻な干ばつ条件下にあったことが報告されており、2002年3月における ^{137}Cs を含む砂塵の多量放出は、干ばつのため植生被覆が脆弱化した草原において強風が吹いたことによると結論付けられた。

春季に日本で ^{137}Cs 降下量が高くなる主要な原因は、以上のような大陸の草原域からの砂塵飛来であると考えられる。しかし、日本国内で発生した局地的な土壌ダストが影響している可能性も指摘されている。従来から広く実施されてきた ^{137}Cs 月間降下量観測の結果から個々のダスト事象の寄与を解明することは困難であり、 ^{137}Cs 降下を正確に評価するためには、黄砂と局地的ダストの両方の影響を判別できる、時間分解能の高い独自の観測が必要とされる。そこで茨城県つくば市において、2007年の春季に ^{137}Cs 降下量の週間観測を実施した。その結果、黄砂由来 ^{137}Cs の寄与は、全観測期間における降下量の67%以上を占め、黄砂飛来が ^{137}Cs 降下の主要な原因であると認められた。また、黄砂由来降下物の単位質量当たりの ^{137}Cs 放射能は 81.8 mBq g^{-1} と高く、黄砂は高い ^{137}Cs 濃度を示すことが明らかになった。

従来、 ^{137}Cs を含む砂塵の主要な供給源は核実験場が立地する中国西部の砂漠域であると考えられてきた。しかし本研究の結果から、供給源としての大陸の半乾燥地の重要性が示された。また、草原土壌から大気への新たな ^{137}Cs 供給プロセスが明らかにされた。これらの成果は大気化学や環境放射能 (放射線) 研究、黄砂研究など幅広い研究分野への学術的貢献として位置づけられるとともに、公衆被曝線量算定への応用面での貢献としても貴重である。