Regular article

Air Pollution with Particulate Matter and Mutagens: Relevance of Asian Dust to Mutagenicity of Airborne Particles in Japan

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To reveal the contamination levels of ambient air with particulate matter and mutagens in Japan and the influence of their long-range transport from the Asian continent, we collected airborne particles at 10 sites, from rural sites to metropolitan areas, in central and western areas of Japan for a year, from July 2008 to June 2009. The fluctuation patterns and levels of airborne particle concentration were similar among the 10 sites, and remarkable increases of the concentrations were seen on a few sampling dates, including March 16 and 17, 2009. Most airborne particles collected at the 10 sites showed mutagenicity toward *Salmonella typhimurium* YG1024 without a mammalian metabolic system (S9 mix) in the Ames test. The mutagenicity levels were quite different among the sampling sites, and the levels seemed to be dependent on the extent of urbanization and industrialization of the subject areas. At Yurihama, a rural site on the west coast of Japan, the mutagenicity level of airborne particles was low, but the particles collected on March 16 and 17, 2009, showed relatively high mutagenicity, >40 revertants/m³, in YG1024 without S9 mix. Airborne particles collected on March 16 and 17, 2009, at most sites

showed relatively high or high mutagenicity, >80 revertants/m³, in YG1024 without S9 mix. High coefficients of correlation were found between the concentration and mutagenicity of airborne particles collected in spring, from March to May, for many sites. The results of back trajectory analysis indicated that air masses for March 16 and 17, 2009, had moved from the Asian continent to Japan. The arrival of Asian dust in central and western areas of Japan on those days was reported by the Japan Meteorological Agency. These results suggest that the arrival of mutagens accompanied by Asian dust could be evidently found at Yurihama on March 2009.

Key words: ambient air, suspended particle, Japan, Asian dust, Ames test

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Introduction

Many epidemiological studies have demonstrated that ambient air pollution is associated with mortality from lung cancer, respiratory diseases, and so forth (1–6). The International Agency for Research on Cancer (IARC) classified outdoor air pollution and particulate matter, a major component of outdoor air pollution, as “carcinogenic to humans (Group 1)” (7). Some particulates occur naturally from volcanoes, forest fires, and so forth. Human activities, such as the burning of fossil fuels in motor vehicles and power plants, also generate particulate matter and emit it into the air. Mutagenic/carcinogenic substances, for example, polycyclic aromatic hydrocarbons (PAHs), are also formed by incomplete combustion of organic material such as fossil fuel and emitted into the air from various anthropogenic sources (8–11). Therefore, the pollution level of ambient air in terms of particulate matter and mutagenic/carcinogenic substances is thought to depend on the extent of urbanization and industrialization of the subject area. On the other hand, it is well known that the long-range transport of air pollutants occurs (12–16). For instance, Katayama *et al.* (12) examined the source-receptor relationship for sulfur in East Asia in December 1995, using a regional chemical transport model coupled with a Regional Atmospheric Modeling System, and estimated that the total contribution of transborder pollution from the Asian continent to sulfur deposition in Japan was almost 75%. This transborder pollution was thought to be caused by the seasonal wind (winter monsoon), which blow northeast to Japan. Moreover, Asian dust (yellow dust or yellow sand) affects much of East Asia, including Japan, sporadically during the spring. Asian dust originates in arid areas in northwestern China and southern Mongolia, such as the Takla Makan and Gobi deserts, and is then carried eastward by Westerlies and seasonal winds and passes over China, Korea, and Japan. In 2009, the total energy consumption in China reached 3.1 billion tons of coal equivalents (17). China has overtaken the United States to become the world’s largest energy user. With the rapid growth of the domestic economy and urbanization in China, the emissions of air pollutants from coal combustion, industrial production, and transport have been increasing at an unprecedented rate over the last decade (18). Therefore, mutagenic/carcinogenic substances originating from these anthropogenic sources may reach other countries by long-range transport via the winter monsoon and Westerlies like Asian dust. However, there are few reports on the long-range transport of mutagenic/carcinogenic substances from the Asian continent to Japan.

In the present study, we collected airborne particles at 10 sites, from rural sites to metropolitan areas, in Japan over a year, from July 2008 to June 2009, and assayed

their mutagenicity by the *Salmonella* Ames test to reveal the contamination levels of the ambient air in Japan with airborne particles and mutagens and the influence of their long-range transport from the Asian continent. The size distributions of particles and 10 PAHs, classified as priority pollutants by the United States Environmental Protection Agency, were also examined for airborne particles collected in Kyoto from January 17 to 21, 2011 (without an Asian dust event), and from May 1 to 6, 2011 (with an Asian dust event). The movements of air masses from which we collected airborne particles were simulated by back trajectory analysis. The influence of the long-range transport of airborne particles and mutagens was also discussed.

Material and Methods

Reagents: Quartz filters were purchased from Pall Life Sciences (Port Washington, NY, USA). Benzo[*a*]pyrene (BaP, CAS 50-32-8), benzo[*b*]fluoranthene (BbF, CAS 205-99-2), benzo[*k*]fluoranthene (BkF, CAS 207-08-9), and indeno[1,2,3-*cd*]pyrene (IcdP, CAS 193-39-5) were purchased from Wako Pure Chemical Industries, Ltd. (Osaka, Japan). Benz[*a*]anthracene (BaA, CAS 56-55-3) and dibenz[*a,h*]anthracene (DahA, CAS 53-70-3) were purchased from Tokyo Chemical Industry Co., Ltd. (Tokyo, Japan). Fluoranthene (FR, CAS 206-44-0) and pyrene (PY, CAS 129-00-0) were purchased from Nacalai Tesque Inc. (Kyoto, Japan). Chrysene (CHR, 218-01-9) and benzo[*ghi*]perylene (BghiP, CAS 191-24-2) were purchased from Sigma-Aldrich Co. LLC (St. Louis, MO, USA). All other chemicals and reagents were of analytical grade.

Sampling of airborne particles and sample preparation: Total suspended particles were collected at 10 sites (Fig. 1), namely, Sasebo City (Nagasaki Prefecture), Dazaifu City (Fukuoka Prefecture), Kitakyushu City (Fukuoka Prefecture), Yurihama Town (Tottori Prefecture), Kobe City (Hyogo Prefecture), Osaka City (Osaka Prefecture), Kyoto City (Kyoto Prefecture), Nagoya City (Aichi Prefecture), Kanazawa City (Ishikawa Prefecture), and Wako City (Saitama Prefecture), located across a wide area of the center and west of Japan. Total suspended particles are thought to be airborne particles with diameters less than 100 μm (19). The characteristics of the sampling sites are shown in Table 1. Kobe, Osaka, and Kyoto constitute the Keihanshin metropolitan area, the second largest metropolitan area in Japan. Nagoya is a core city of the Chukyo metropolitan area, the third largest metropolitan area. Wako is located in the Kanto metropolitan area, the largest metropolitan area. Kitakyushu is a core city of the Kitakyushu-Fukuoka metropolitan area, the fourth largest metropolitan area in Japan. Dazaifu is located 16 km southeast of Fukuoka City, another core city of the Kitakyushu-Fukuoka metropolitan area.

Fig. 1. Sampling sites of airborne particles.

Table 1. Characteristics of sampling sites

Sampling site	Population	Population density (/km ²)	Metropolitan area	Characteristic of sampling site (Major industry)
Sasebo	258,430	606	—	Countryside 10 km away from harbor (Shipbuilding)
Dazaifu	71,200	2,410	Kitakyushu-Fukuoka	Countryside 16 km away from Fukuoka commercial area (Sightseeing, farming)
Kitakyushu	971,931	1,990	Kitakyushu-Fukuoka	Residential area near industrial area (Heavy industry, sightseeing)
Yurihama	16,884	217	—	Countryside far from any metropolitan area (Sightseeing, farming)
Kobe	1,542,230	2,790	Keihanshin	Harbor near commercial area (Heavy industry, shipbuilding, sightseeing)
Osaka	2,678,549	12,010	Keihanshin	Commercial area (Heavy industry, sightseeing)
Kyoto	1,473,069	1,780	Keihanshin	Residential area (Sightseeing, traditional industry, farming)
Nagoya	2,268,423	6,950	Chukyo	Industrial area (Heavy industry)
Kanazawa	463,679	991	—	Mountainside 6 km away from commercial area (Sightseeing, traditional industry, farming, fishing)
Wako	81,315	7,370	Kanto	Rural area next to Tokyo Prefecture (Heavy industry)

Therefore, Dazaifu was thought to be influenced by Fukuoka. Populations of Sasebo and Kanazawa are about 258,000 and 464,000, respectively, and are considered as medium-sized cities. Neither of them belongs to any metropolitan area. The sampling site in Sasebo is lo-

cated near a harbor, and that in Kanazawa is 6 km away from the commercial area and surrounded by mountains. Yurihama is in the countryside. Sampling sites in Yurihama and Kanazawa have few large emission sources of air pollutants.

Fig. 2. Concentration of airborne particles collected at the 10 sites from July 2008 to June 2009. Sampling dates when collection was not performed are shaded.

The sampling periods were as follows: July 14–17, August 4–7, September 8–11, October 6–9, November 10–13, and December 8–11 in 2008; and January 5–8, February 20–23, March 9–12, 16–19, April 6–9, 27–30, May 11–14, 25–28, and June 15–18 in 2009. In this study, seasons were defined as follows: spring months are March, April, and May; summer months are June, July, and August; autumn months are September, October, and November; and winter months are December, January, and February.

The sampling of airborne particles started at about 10 AM (Japan Standard Time, JST) and finished at about 10 AM the next day at every sampling site. The airborne particles were collected on quartz filters at a flow rate of 700–1400 L/min with high-volume air samplers (Shibata Scientific Technology Ltd., Soka, Japan, or Kimoto Electric Co. Ltd., Osaka, Japan) for about 24 h. Total air volume was in the range of 900–1600 m³. The filters were weighted before and after sampling after being humidified at 50% relative humidity with sodium hydrogen sulfate solution at 20°C. To calculate the concentration of airborne particles, the weight of airborne particles collected on a filter was divided by the volume of the air that had passed through the filter.

Extracts of airborne particles were prepared as described previously (20). Forty percent of the filter (corresponding to 360–640 m³ air) was cut into small pieces and extracted with 100 mL of methanol using an ultrasonic apparatus for 20 min. All airborne particle extracts were filtered through No. 5C filter papers (Advantec Toyo, Tokyo, Japan) and 75 ml of filtrate was evaporated to dryness.

Mutagenic assay: The extracts of airborne particles were dissolved in dimethyl sulfoxide and assayed by the preincubation method (21) using *Salmonella typhimurium* YG1024 (22) and YG1029 (22) with and without S9 mix. YG1024 and YG1029 were kindly pro-

vided by Dr. Nohmi from the National Institute of Health. YG1024 and YG1029 are *O*-acetyltransferase-overproducing derivatives of *Salmonella typhimurium* TA98 and TA100, respectively, and are sensitive to mutagens that require metabolic activation by *O*-acetyltransferase, such as nitroaromatic compounds. Nitroaromatic compounds, such as 1-nitropyrene, are representative environmental mutagens. YG1024 and YG1029 were used in this study because of the sensitivity to these environmental mutagens. The S9 mix contained 0.05 mL of S9, prepared from livers of male Sprague-Dawley rats treated with phenobarbital and β -naphthoflavone, in a total volume of 0.5 mL. The mutagenic potencies of samples were calculated from linear portions of the dose-response curves obtained with three or four doses and duplicated plates at each dose. The

Table 3. Concentrations of airborne particles at the 10 sites in the four seasons and annually

Period	Concentration of airborne particles ($\mu\text{g}/\text{m}^3$)				
	Sasebo	Dazaifu	Kitakyushu	Yurihama	Kobe
Summer	42 $\pm$ 12	40 $\pm$ 9	42 $\pm$ 15	33 $\pm$ 3	33 $\pm$ 8
Autumn	37 $\pm$ 16	34 $\pm$ 10	46 $\pm$ 13	30 $\pm$ 10	36 $\pm$ 15
Winter	57 $\pm$ 49	56 $\pm$ 40	64 $\pm$ 53	42 $\pm$ 32	48 $\pm$ 23
Spring	64 $\pm$ 25	63 $\pm$ 28	70 $\pm$ 34	69 $\pm$ 58	56 $\pm$ 26
Year	51 $\pm$ 28	50 $\pm$ 27	59 $\pm$ 34	46 $\pm$ 40	45 $\pm$ 22

Period	Concentration of airborne particles ($\mu\text{g}/\text{m}^3$)					
	Osaka	Kyoto	Nagoya	Kanazawa	Wako	10 sites
Summer	38 $\pm$ 4	43 $\pm$ 13	52 $\pm$ 16	26 $\pm$ 6	41 $\pm$ 20	39 $\pm$ 13
Autumn	40 $\pm$ 10	64 $\pm$ 46	52 $\pm$ 18	47 $\pm$ 25	37 $\pm$ 9	42 $\pm$ 22
Winter	51 $\pm$ 26	59 $\pm$ 46	61 $\pm$ 38	78 $\pm$ 54	48 $\pm$ 24	56 $\pm$ 39
Spring	70 $\pm$ 39	71 $\pm$ 44	71 $\pm$ 38	62 $\pm$ 38	75 $\pm$ 39	67 $\pm$ 37
Year	56 $\pm$ 32	62 $\pm$ 41	62 $\pm$ 32	55 $\pm$ 39	58 $\pm$ 34	55 $\pm$ 34

Numbers express mean values $\pm$ standard deviation.

Table 2. The total sampling days and dates of high airborne particle concentration ($> 100 \mu\text{g}/\text{m}^3$)

Sampling site	Total sampling days	Sampling date of high airborne particle concentration												Number of high-concentration days
		2008						2009						
		Jul.	Aug.	Sep.	Oct.	Nov.	Dec.	Jan.	Feb.	Mar.	Apr.	May	Jun.	
Sasebo	52	—	—	—	—	—	—	—	<u>20</u>	—	7	—	—	2
Dazaifu	54	—	—	—	—	—	—	—	<u>20, 21</u>	<u>17</u>	7, 8	—	—	5
Kitakyushu	52	—	—	—	—	—	10, 11	—	<u>20</u>	<u>17</u>	7, 8, 9	—	—	7
Yurihama	46	—	—	—	—	—	—	—	<u>20</u>	<u>16, 17</u>	—	—	—	3
Kobe	51	—	—	—	—	—	—	—	—	—	—	13	—	1
Osaka	52	—	—	—	—	—	11	—	—	<u>16, 17, 18</u>	—	—	—	4
Kyoto	58	—	—	9, 10, 11	6	—	10	—	23	<u>16, 17, 18</u>	7, 9	—	—	11
Nagoya	52	—	—	—	—	—	10, 11	—	—	<u>16, 17</u>	—	—	—	4
Kanazawa	58	—	—	—	—	12	9, 10, 11	—	—	<u>16, 17</u>	—	—	—	6
Wako	50	—	—	—	—	—	—	—	—	<u>16, 18</u>	—	11, 12, 14	—	5

The dates when Asian dust arrived as reported by the Japan Meteorological Agency (25) are underlined.

Fig. 3. Mutagenicities of airborne particles collected at the 10 sites from July 2008 to June 2009. Sampling dates when collection was not performed are shaded. The mean values of negative control were as follows: 43 revertants/plate (YG1024, -S9 mix), 133 revertants/plate (YG1024, +S9 mix), 160 revertants/plate (YG1029, -S9 mix), 165 revertants/plate (YG1029, +S9 mix). The mean values of positive control were as follows: 353 revertants/plate (YG1024, -S9 mix, 1-nitropyrene 0.01 $\mu\text{g}/\text{plate}$), 473 revertants/plate (YG1024, +S9 mix, acetylaminofluorene 0.02 $\mu\text{g}/\text{plate}$), 584 revertants/plate (YG1029, -S9 mix, furlfuramide 0.02 $\mu\text{g}/\text{plate}$), 1163 revertants/plate (YG1029, +S9 mix, BaP 2 $\mu\text{g}/\text{plate}$).

slope of each dose-response curve was adopted as the mutagenic potency. When the samples induced twofold increases over the average of spontaneous revertants and showed well-behaved concentration-response patterns, the samples were judged as positive. As positive controls, BaP and 2-acetylaminofluorene were used in the presence of S9 mix, and 1-nitropyrene and furlfuramide were utilized in the absence of S9 mix

Sampling of size-fractionated airborne particles and analysis of PAHs: Size-fractionated airborne particles, namely, < 1.1, 1.1–2.0, 2.0–3.3, 3.3–7.0, and > 7.0 μm , were collected in Kyoto on quartz filters at a flow rate of 566 L/min with an Andersen-type high-volume air sampler (Shibata Scientific Technology Ltd., Soka, Japan) from January 17 to 21, 2011 (without an Asian dust event), and from May 1 to 6, 2011 (with an

Fig. 3. Continued

Asian dust event). Extracts of airborne particles were prepared as described previously (23). The filter with particle sizes smaller than $1.1\ \mu\text{m}$ (corresponding to 407.5 or 509.4 m³) was cut into small pieces and extracted with 100 mL of methanol twice using an ultrasonic apparatus for 15 min each time. The filters with particle sizes larger than $1.1\ \mu\text{m}$ (corresponding to 815.0 or 1018.8 m³) were cut into small pieces and similarly extracted with 200 mL of methanol and 100 mL of it. All extracts were filtered through No. 5C filter papers and the filtrate was evaporated to dryness. Extracts were dis-

solved in 10 mL of ethanol and 50 μL of the solutions were injected into a Wakosil-PAHs column (4.6 mm $\times$ 250 mm, Wako Pure Chemical Industries, Ltd.) for HPLC. The elution was performed with the following gradient system using solution A (80% methanol) and solution B (acetonitrile): 0–20 min, 10–75% solution B (linear gradient with solution A); 20–30 min, 75% solution B (25% solution A), at a flow rate of 1 mL/min. The eluate was monitored using a fluorescence detector (RF-20AXS, Shimadzu, Kyoto, Japan) with the following excitation (Ex) and emission (Em) wavelengths:

Fig. 3. Continued

0–11.5 min, Ex 250 nm, Em 420 nm; 11.5–18.75 min, Ex 270 nm, Em 400 nm; 18.75–23.3 min, Ex 296 nm, Em 410 nm; 23.3–30 min, Ex 300 nm, Em 500 nm. Contents of PAHs in samples were estimated from standard curves obtained from various doses of the authentic compounds.

Statistical analysis: Coefficients of correlation between the concentration and mutagenicity of airborne particles for each sampling site were calculated using Microsoft Office Excel 2013.

Back trajectory analysis: Back trajectory analysis

was performed for each collection date and each sampling site with HYSPLIT (2010) provided by the National Oceanic and Atmospheric Administration (NOAA) of the United States of America (24). The start time of analysis was set at 10 PM (Japan Standard Time), namely, 1 PM UTC (Coordinated Universal Time), and the height was set at 1500 m. Back trajectory analysis was calculated using model vertical velocity and the trajectory duration was 96 h.

Fig. 3. Continued

Results

Daily and seasonal variation of the concentration of airborne particles: Concentrations of airborne particles collected at the 10 sites are shown in Fig. 2. Numbers of sampling days at the 10 sites ranged from 46 to 58 days. The numbers of sampling days at each site and sampling dates when the concentrations of airborne particles were higher than $100 \mu\text{g}/\text{m}^3$ are listed in Table 2. The particle concentrations at each site were about $50 \mu\text{g}/\text{m}^3$ or less on most sampling dates. Very high particle concentrations ($>150 \mu\text{g}/\text{m}^3$) were found at 5 and 6

sites on March 16 and 17, respectively. At 4 sites, namely, Yurihama, Osaka, Kyoto, and Nagoya, particle concentrations were the highest, from 177 to $227 \mu\text{g}/\text{m}^3$ air, on March 16, 2009. At Kanazawa, the second highest concentration of airborne particles was seen on March 16. On December 10 and 11, 2008, and February 20 and April 7, 2009, high particle concentrations ($>100 \mu\text{g}/\text{m}^3$) were seen at 4 sites on each day.

The mean values of airborne particle concentrations at the 10 sites in each season and year are shown in Table 3. The highest concentrations of airborne parti-

Fig. 3. Continued

cles were observed in spring at 9 sites and the concentrations ranged from 63 to 75 $\mu\text{g}/\text{m}^3$. The second highest mean values were found in winter at 8 sites (from 42 to 64 $\mu\text{g}/\text{m}^3$). The lowest concentration of airborne particles was detected in summer or autumn at 5 sites each.

Daily and seasonal variation of the mutagenicity of airborne particles: Figure 3 shows the mutagenicities of organic extracts of airborne particles collected at the 10 sites in YG1024 and YG1029 with and without S9 mix. Most samples from the 10 sites were mutagenic in YG1024 with and without S9 mix, and most samples

showed higher activities without S9 mix than with it. Although many samples showed mutagenicity in YG1029, the potencies were generally lower than those in YG1024. Mutagenicity levels of airborne particles (the highest mutagenicities and numbers of samples that showed potent mutagenicity) were quite different among the sampling sites. At Yurihama, the mutagenicity level of airborne particles was low, but 3 samples, collected on December 10, 2008, and March 16 and 17, 2009, showed relatively high mutagenicity, between 40 and 80 revertants/ m^3 , in YG1024 without S9 mix. The sam-

Table 4. Sampling dates of airborne particles inducing more than 40 revertants/m³ in YG1024

Sampling site	2008						2009						Number of days
	Jul.	Aug.	Sep.	Oct.	Nov.	Dec.	Jan.	Feb.	Mar.	Apr.	May	Jun.	
without S9 mix													
Sasebo	—	7	—	7	—	—	5, 6, 8	—	10, 16	7	—	—	8
Dazaifu	—	—	—	7	—	10, 11	5, 6, 7, 8	<u>20, 21</u>	16	6, 7, 8, 9	14	—	15
Kitakyushu	—	4, 7	10, 11	7, 8, 9	—	10, 11	6, 7, 8	<u>20, 21, 22</u>	9, 10, <u>17</u>	7, 8	—	—	20
Yurihama	—	—	—	—	—	10	—	—	<u>16, 17</u>	—	—	—	3
Kobe	14, 15	7	10, 11	9	13	8, 9, 10, 11	7, 8	—	9	6, 7, 8, 9, 30	—	—	19
Osaka	—	6, 7	10, 11	6, 7, 8, 9	11, 13	8, 9, 10, 11	7, 8	<u>20, 21</u>	9, 12, <u>16, 17, 18</u>	6, 7, 8, 9, 28	11, 12, 26, 28	—	32
Kyoto	—	—	—	7	13	8, 10, 11	7	—	<u>12, 16, 17, 18, 19</u>	—	—	—	11
Nagoya	—	4, 5, 6, 7	10	7, 8, 9	11, 13	8, 9, 10, 11	7, 8	21, 23	9, <u>12, 16, 17</u>	6, 7, 8, 9	11	—	27
Kanazawa	16	—	11	—	—	—	—	—	—	—	—	—	2
Wako	15, 16	5	9	—	11, 12, 13	8, 9, 11	5, 7, 8	22	9, 10, 12, 16, <u>17, 18</u>	7, 8	11	18	24
with S9 mix													
Sasebo	—	—	—	7	—	—	6	—	10, 19	7	—	—	5
Dazaifu	—	—	—	7, 9	—	8, 10	5, 6, 7, 8	—	—	7, 8, 9	—	—	11
Kitakyushu	—	—	—	7, 8, 9	13	10	6, 8	—	9, 10, 11	7, 8, 9	26, 27	—	15
Yurihama	—	—	—	—	—	—	—	—	—	—	—	—	0
Kobe	—	4	—	9	13	8, 9, 10	—	—	9	6, 7, 8	—	—	10
Osaka	—	—	10, 11	7, 8, 9	11, 13	8, 9, 10, 11	7	—	9, <u>16, 17, 18</u>	6, 7, 8	11, 26, 28	—	22
Kyoto	—	—	—	7	13	8, 10, 11	7	—	9, <u>16, 17</u>	—	—	—	9
Nagoya	—	—	—	7, 8, 9	11, 12, 13	8, 9, 10, 11	7, 8	21	9, 12, <u>16, 17</u>	6, 7, 9, 28	11, 26, 28	—	24
Kanazawa	—	—	—	—	—	—	—	—	—	—	—	—	0
Wako	15, 16	—	—	—	11, 13	8, 9, 10	7, 8	22	9, 10, 12, 16, 18	—	—	—	15

The dates when Asian dust arrived as reported by the Japan Meteorological Agency (25) are underlined.

pling dates of airborne particles that induced more than 40 revertants/m³ in YG1024 without S9 mix are shown in Table 4. At Kitakyushu, Kobe, Osaka, Nagoya, and Wako, about 20 samples or more induced >40 revertants/m³ in YG1024 without S9 mix. At Sasebo, Dazaifu, and Kyoto, the numbers of sampling dates of particles inducing >40 revertants/m³ were moderate, from 8 to 15 days. Only 2 samples from Kanazawa induced >40 revertants/m³. As shown in Fig. 3, many samples collected at Osaka and Nagoya showed high mutagenicity, inducing more than 80 revertants/m³, in YG1024 without S9 mix. Airborne particles collected at 7 sites on March 16, 2009, induced >40 revertants/m³, and those from Sasebo, Osaka, Kyoto, and Nagoya showed high mutagenicity, >80 revertants/m³. Similarly, airborne particles collected at 6 sites on March 17, 2009, induced >40 revertants/m³, and those from Osaka and Nagoya showed high mutagenicity. Relatively high or high mutagenicities were detected for 7 samples collected on December 10 and 11, 2008.

Table 5 shows seasonal and annual mean values of the mutagenicities of airborne particles from the 10 sites in YG1024 and YG1029 with and without S9 mix. The highest annual means were observed in YG1024 without S9 mix at 9 sites. The annual means of the 10 sites in YG1024 without S9 mix were quite different among the sampling sites. Low annual means were seen at Kanazawa and Yurihama, 12.2 and 13.1 revertants/m³, respectively, and high means were observed at Kitakyushu,

Kobe, Osaka, Nagoya, and Wako, from 38.1 to 60.9 revertants/m³. For the seasonal means in YG1024 without S9 mix, the highest values were observed in winter at 9 sites, and the second highest values were found in autumn and spring at 5 and 3 sites, respectively.

Correlation between the concentration and mutagenicity of airborne particles: Coefficients of correlation between the concentration and mutagenicity of airborne particles at each sampling site in each season and year are shown in Table 6. In YG1024 without S9 mix, a positive correlation (correlation coefficients were larger than 0.4) were observed at 8 sites in spring, and correlation coefficients were larger than 0.6 at 6 sites, namely, Dazaifu, Yurihama, Osaka, Kyoto, Nagoya, and Kanazawa. Figure 4 shows the correlation between the concentration and mutagenicity of airborne particles collected at these 6 sites in spring. The correlation coefficient for Yurihama was especially large, 0.909. In autumn and winter, large correlation coefficients (>0.6) were found at 4 sites, namely, Dazaifu, Kitakyushu, Kobe, and Osaka, and 2 sites, namely, Osaka and Nagoya, respectively. The correlation coefficient for Yurihama in year was 0.668. In YG1024 with S9 mix, high correlation coefficients were observed at 4 sites in autumn.

Back trajectory analysis: Figure 5 shows the results of back trajectory analysis of the air masses of 8 sites for March 16, 2009. Dazaifu and Kitakyushu are located in Fukuoka prefecture. The distance between

Table 5. Mutagenicity of airborne particles at the 10 sites in the four seasons and annually

Period	Sasebo				Dazaifu				Kitakyushu			
	YG1024		YG1029		YG1024		YG1029		YG1024		YG1029	
	−S9 mix	+S9 mix	−S9 mix	+S9 mix	−S9 mix	+S9 mix	−S9 mix	+S9 mix	−S9 mix	+S9 mix	−S9 mix	+S9 mix
Summer	16.1±19.5	6.4± 5.9	0.0± 0.0	0.4± 1.3	15.1± 9.2	9.3± 8.8	4.0± 4.6	4.3± 8.2	35.6±38.9	12.5± 7.8	6.6± 7.4	6.7± 8.6
Autumn	14.8±10.7	14.8±18.3	4.0± 7.2	6.1± 8.5	24.7±16.1	20.3±18.8	4.1± 5.6	5.4± 5.3	44.0±25.7	47.8±39.8	14.5±13.9	24.7±19.7
Winter	48.6±39.4	21.4±16.6	13.9± 8.4	5.8± 7.4	60.5±31.4	39.4±23.8	8.6± 3.5	1.0± 3.2	57.9±18.3	30.3±28.7	14.5± 9.0	23.6±31.6
Spring	23.5±23.1	14.9±15.9	3.9± 5.3	4.9± 7.5	28.9±23.3	20.1±21.4	6.0± 6.2	6.9± 7.9	28.1±23.0	28.4±27.4	7.2± 8.0	12.2±16.5
Year	23.6±25.4	13.9±15.3	4.6± 7.1	4.2± 7.0	31.3±26.3	21.7±21.3	5.7± 5.4	4.8± 6.9	38.1±27.5	30.8±30.4	10.0±10.2	16.2±20.4

Period	Yurihama				Kobe				Osaka			
	YG1024		YG1029		YG1024		YG1029		YG1024		YG1029	
	−S9 mix	+S9 mix	−S9 mix	+S9 mix	−S9 mix	+S9 mix	−S9 mix	+S9 mix	−S9 mix	+S9 mix	−S9 mix	+S9 mix
Summer	9.3± 7.4	4.0± 5.4	1.1± 2.8	2.0± 3.4	29.9±22.8	13.0±14.4	5.3± 4.9	6.7± 7.4	72.2±55.0	20.5±11.8	13.2±14.8	4.8± 6.2
Autumn	7.7± 4.5	3.1± 3.4	0.9± 3.0	1.1± 3.8	39.4±29.4	23.7±25.6	14.1±10.0	19.4±20.2	50.6±24.6	40.0±25.5	19.4±14.9	29.5±24.1
Winter	21.8± 9.4	9.8± 9.2	5.7± 6.1	1.4± 3.5	53.9±26.9	34.0±27.5	17.3± 8.4	11.6±10.7	69.9±36.3	43.1±34.6	22.9±14.1	13.2±16.3
Spring	12.2±16.1	5.1± 7.5	1.6± 4.2	1.8± 3.9	34.0±26.6	31.4±30.7	12.3±11.0	16.4±17.0	59.6±38.8	51.2±40.5	19.7±13.6	24.3±20.8
Year	13.1±12.0	5.6± 7.2	2.4± 4.7	1.5± 3.6	38.4±27.1	26.1±26.5	12.1± 9.8	13.7±15.1	60.9±36.4	44.4±34.9	19.9±13.9	21.5±20.9

Period	Kyoto				Nagoya				Kanazawa			
	YG1024		YG1029		YG1024		YG1029		YG1024		YG1029	
	−S9 mix	+S9 mix	−S9 mix	+S9 mix	−S9 mix	+S9 mix	−S9 mix	+S9 mix	−S9 mix	+S9 mix	−S9 mix	+S9 mix
Summer	16.5± 6.7	9.1± 6.2	1.7± 3.2	6.2± 5.2	36.9±31.6	13.0±11.4	5.3± 5.2	10.3± 9.1	12.4±11.3	1.4± 1.9	0.0± 0.0	0.4± 1.5
Autumn	26.7±16.5	20.7±17.6	6.9± 6.0	12.0±11.3	46.8±26.7	58.8±47.5	22.7±23.1	38.1±35.9	11.4± 9.8	4.6± 2.5	0.8± 2.7	1.4± 2.5
Winter	40.0±23.7	29.8±28.7	6.7±10.6	6.3± 6.5	59.1±28.6	63.7±57.6	21.0±15.2	25.9±23.9	14.8± 8.2	4.2± 4.3	0.7± 2.3	0.0± 0.0
Spring	25.3±22.0	22.4±21.0	6.2± 7.3	12.4±13.5	35.1±28.0	37.0±26.3	9.7± 9.5	23.0±21.5	11.2± 9.4	6.4± 7.8	1.3± 2.7	2.3± 5.2
Year	26.6±20.0	20.7±20.7	5.5± 7.3	9.9±10.8	43.2±29.2	44.1±41.6	14.4±15.6	25.1±25.5	12.2± 9.5	4.6± 5.8	0.8± 2.3	1.3± 3.7

Period	Wako				10 sites			
	YG1024		YG1029		YG1024		YG1029	
	−S9 mix	+S9 mix	−S9 mix	+S9 mix	−S9 mix	+S9 mix	−S9 mix	+S9 mix
Summer	32.1±17.0	27.8±32.5	10.9± 9.7	21.6±19.2	24.0±25.2	10.8±14.2	4.0± 6.5	6.0± 9.8
Autumn	33.4±16.3	29.8±20.6	14.6±10.3	28.8±18.3	29.8±24.1	26.4±30.6	10.0±13.4	16.2±21.5
Winter	58.9±35.1	48.3±35.1	10.5± 7.0	18.1±18.1	48.4±31.5	32.9±34.3	12.2±11.4	10.7±17.2
Spring	32.7±25.0	31.1±30.8	14.7±15.1	24.4±23.2	29.7±27.7	25.6±28.4	8.5±10.8	13.4±17.6
Year	38.4±27.1	34.1±31.0	13.1±12.0	23.1±20.5	32.5±28.6	24.6±29.0	8.8±11.2	12.1±17.6

Numbers express mean values (rev./m³)±standard deviation.

the sampling sites in these cities was 43 km, so that trajectories from these sites were quite similar. Trajectories from 7 sites, namely, Sasebo, Dazaifu, Kitakyushu, Yurihama, Osaka, Kanazawa, and Wako, indicate that air masses at these sites had transferred from the northwest of China or southern Mongolia, namely, the Gobi desert, without a large detour. For Kyoto and Nagoya, the trajectories suggest that air masses had transferred from the central or eastern areas of Mongolia to Zhejiang province and its environs in China, and then been carried to each site in Japan. For March 17, 2009, the trajectories of air masses for each site indicate that air masses of all sites were transferred through the Gobi desert and reached each site without an obvious detour

(data not shown). Airborne particles were not collected in Kobe on those days.

Size distributions of airborne particles and PAHs:

Figure 6(A) shows the size distributions of airborne particles collected in Kyoto from January 17 to 21, 2011 (without an Asian dust event) and from May 1 to 6, 2011 (with an Asian dust event). The size distributions are shown as the ratios of weights of particles classified into 5 types, namely, <1.1, 1.1–2.0, 2.0–3.3, 3.3–7.0, and >7.0 μm. For the period without an Asian dust event (January 17–21, 2011), the smallest particles (<1.1 μm) were the most abundant and accounted for 70% of the total. Large size fractions of >7.0 and 3.3–7.0 μm accounted for only 8% and 14%, respec-

Table 6. Coefficients of correlation between the concentration and mutagenicity of airborne particles

Sampling site	YG1024, –S9 mix					YG1024, +S9 mix				
	Summer	Autumn	Winter	Spring	Year	Summer	Autumn	Winter	Spring	Year
Sasebo	–0.098	0.408	–0.124	0.285	0.153	–0.434	0.372	–0.373	0.239	0.083
Dazaifu	0.229	0.730	–0.209	0.644	0.292	0.108	0.792	–0.257	0.602	0.272
Kitakyushu	0.351	0.774	0.390	0.485	0.287	0.549	0.627	0.320	0.320	0.263
Yurihama	0.346	0.455	0.496	0.909	0.668	0.297	0.704	0.048	0.416	0.249
Kobe	–0.293	0.786	0.294	0.530	0.374	–0.373	0.407	0.413	0.373	0.410
Osaka	–0.618	0.718	0.649	0.709	0.553	0.492	0.649	0.558	0.408	0.473
Kyoto	–0.180	–0.158	0.053	0.749	0.353	–0.471	–0.349	–0.015	0.589	0.226
Nagoya	–0.495	0.329	0.600	0.828	0.481	–0.249	0.108	0.527	0.501	0.316
Kanazawa	0.285	–0.004	0.394	0.669	0.378	0.431	0.296	0.665	0.330	0.415
Wako	0.553	0.171	0.241	0.302	0.165	0.644	0.166	0.357	–0.045	0.084

Sampling site	YG1029, –S9 mix					YG1029, +S9 mix				
	Summer	Autumn	Winter	Spring	Year	Summer	Autumn	Winter	Spring	Year
Sasebo	NA	0.190	–0.293	0.379	0.121	–0.064	0.102	–0.341	0.329	0.082
Dazaifu	0.503	0.802	0.048	0.488	0.396	0.306	0.497	0.274	0.588	0.344
Kitakyushu	0.775	0.279	–0.175	0.274	0.081	0.623	–0.045	0.706	0.068	0.286
Yurihama	–0.170	0.188	0.314	0.931	0.464	0.049	0.188	–0.044	0.534	0.287
Kobe	0.334	0.363	0.244	0.394	0.369	0.261	0.257	0.378	0.450	0.368
Osaka	0.086	0.179	0.578	0.263	0.270	0.420	0.342	0.701	0.022	0.165
Kyoto	0.058	–0.132	–0.047	0.602	0.274	–0.272	–0.173	0.080	0.363	0.211
Nagoya	–0.332	0.227	0.270	0.510	0.190	–0.218	0.174	0.507	0.555	0.338
Kanazawa	NA	–0.037	0.057	0.271	0.198	–0.064	0.861	NA	0.181	0.182
Wako	0.541	–0.145	0.579	0.308	0.339	0.537	–0.082	0.521	0.182	0.233

NA: Not Available.

tively. In contrast, large size fractions were the major fractions for the period including an Asian dust event, and >7.0 and $3.3\text{--}7.0\text{ }\mu\text{m}$ size fractions accounted for 25 and 29%, respectively. The smallest size fraction was 18%. Figure 6(B) shows the size distributions of PAHs in the airborne particles collected in two periods described above. Ten PAHs, namely, BaP, BbF, BkF, IcdP, BaA, DahA, FR, PY, CHR, and BghiP, were analyzed by HPLC using a fluorescence detector. PAHs were detected at retention times as follows: FR, 9.3 min; PY, 10.2 min; BaA, 13.2 min; CHR, 14.3 min; BbF, 16.6 min; BkF, 18.2 min; BaP 19.6 min; DahA, 21.7 min; BghiP, 22.7 min; and IcdP, 23.8 min. The size distributions of PAHs are shown as ratios of the total weights of 10 PAHs detected in each size fraction. As shown in Fig. 6(B), 78% and 10% of total PAHs were detected in small size fractions of <1.1 and $1.1\text{--}2.0\text{ }\mu\text{m}$, respectively, for the period without an Asian dust event. Similarly, 61 and 18% of PAHs were found in small size fractions of <1.1 and $1.1\text{--}2.0\text{ }\mu\text{m}$, respectively, for the period including an Asian dust event.

Discussion

In the present study, we collected airborne particles at 10 sites in central and western areas of Japan. These 10 sites can be classified into 3 groups: metropolitan areas, moderately urbanized sites, and rural sites. Kitakyushu,

Kobe, Osaka, Kyoto, Nagoya, and Wako are in metropolitan areas, and Sasebo and Dazaifu are moderately urbanized sites. Yurihama is a rural site. Although Kanazawa is a medium-sized city, the sampling site is far from the commercial area and has few large emission sources of air pollution. Therefore the sampling site in Kanazawa is thought to be a rural site. The concentrations of airborne particles at the 10 sites were generally about $50\text{ }\mu\text{g}/\text{m}^3$ or less, and simultaneous increases of the particle concentration were found at several sites in winter and spring (Fig. 2 and Table 2). These results suggest that the fluctuations of airborne particle concentrations among the 10 sites throughout the period in this study were similar. At many sampling sites, the particle concentrations were especially high on March 16 and/or 17, 2009. On those days, Asian dust was found at many sampling sites by monitoring conducted by the Japan Meteorological Agency (25). The increased particle concentrations found in March are thought to have been caused by Asian dust. Relatively high concentration of airborne particles and mutagenicity were found at many sites on December 10 and 11, 2008, as shown in Fig. 2 and Fig. 3. Relatively high concentration of a sulfate ion were found in ambient air at those sites on those days (data not shown). A sulfate ion is a major component of fine particles, such as $\text{PM}_{2.5}$, and it was reported that the concentration of a sulfate

Fig. 4. Correlation between the concentration and mutagenicity of airborne particles collected at Dazaifu, Yurihama, Osaka, Kyoto, Nagoya and Kanazawa in spring 2009.

ion in ambient air was mainly effected by long-range transport of air pollution from the Asian continent (19,26–28). The relatively high concentration of airborne particles and mutagenicity on December 10 and 11, 2008, may be caused by long-range transport of air pollutant. Most samples collected at the 10 sites showed the highest mutagenicity in YG1024 without S9 mix, and the potencies differed among the sampling sites (Fig. 5). The numbers of sampling dates of particles that showed relatively high or high mutagenicity were large for Kitakyushu, Kobe, Osaka, Nagoya, and Wako, moderate for Kyoto, Sasebo, and Dazaifu, and small for Yurihama and Kanazawa (Table 4). The annual means of mutagenic potencies in YG1024 without S9 mix were high for Kitakyushu, Kobe, Osaka, Nagoya, and Wako,

moderate for Sasebo, Dazaifu, and Kyoto, and low for Yurihama and Kanazawa (Table 5). These results suggested that contamination levels of ambient air with mutagens were as follows: metropolitan areas > moderately urbanized sites > rural sites. The correlation coefficients between the concentration and mutagenicity of airborne particles collected at the 10 sites are shown in Table 6. In YG1024 without S9 mix, a positive correlation was observed in spring at most sites, and correlation coefficients were larger than 0.6 at 6 sites, namely, Dazaifu, Yurihama, Kobe, Osaka, Kyoto, Nagoya, and Kanazawa. The correlation coefficient for Yurihama was especially large, 0.909. On the other hand, high correlation coefficients, namely, >0.6 , were observed at only 2 sites in winter. These results suggest that the

Fig. 5. Back trajectories of air masses of 8 sites from March 16, 2009, for 96 h. Back trajectory analysis was performed with HYSPLIT (2010) provided the NOAA of the USA (24).

Fig. 6. Size distributions of airborne particles (A) and PAHs (B) collected in Kyoto.

mutagenic potency of ambient air was associated with the concentration of airborne particles in spring, and potent mutagenicities were detected for the samples collected on days when the particle concentrations were high, such as March 16 or 17, 2009. The results of back trajectory analysis indicated that air masses for those days moved from the Asian continent to each sampling site in Japan (Fig. 5). This suggests that airborne particles including Asian dust and mutagens arrived in Japan in March 2009, and long-range transport of mutagens was evident at Yurihama, which has few large emission sources of air pollutants around the sampling site.

Size distributions of airborne particles and 10 PAHs, representative air pollutants formed by incomplete combustion of organic matter such as fossil fuels, were examined for airborne particles collected in Kyoto for two periods with and without an Asian dust event. As shown in Fig. 6, the ratios of large size particles (>7.0 and $3.3\text{--}7.7\text{ }\mu\text{m}$) were high among airborne particles for the period including an Asian dust event, but PAHs were mainly distributed in the smallest size fraction ($<1.1\text{ }\mu\text{m}$) for both periods. It is reported that the mass concentration of airborne particles during Asian dust event was predominated by coarse particles (29–31), and the result in this study was consistent with these findings. There are some reports on the size distribution of PAHs in airborne particles, and PAHs were mainly detected in

small size fractions (32–34). These results suggest that a large number of coarse particles were transported as Asian dust and that air pollutants formed from incomplete combustion, such as PAHs, mainly existed among fine particles for periods with and without an Asian dust event. However, there are few reports on the size distribution of mutagens in airborne particles for periods of an Asian dust event. Long-range transport of fine particles, such as PM_{2.5}, has recently been reported (35). Some fine particles are formed by combustion of organic matter such as fossil fuels and reactions of primary gases in ambient air. Further study is necessary to clarify whether air pollutants are attached to fine particles of Asian dust and transported to Japan. In addition, it is important to clarify whether mutagenic/carcinogenic compounds are transported as fine particles emitted from anthropogenic sources with long-range transport by the winter monsoon.

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