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Articles

A Database of Hourly Atmospheric Concentrations of Radiocesium (^{134}Cs and ^{137}Cs) in Suspended Particulate Matter Collected in March 2011 at 99 Air Pollution Monitoring Stations in Eastern Japan

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In this study, we compiled a comprehensive dataset of hourly atmospheric ^{134}Cs and ^{137}Cs radioactivity concentrations in suspended particulate matter (SPM; aerosols less than 10 μm in diameter) that was systematically collected from March 12 to 23, 2011, in eastern Japan. In Japan, mass concentrations of SPM are measured using samples automatically collected on filter tapes at the air quality monitoring stations managed by the local governments. Most of the SPM monitoring stations in eastern Japan were operated during and/or after the Great East Japan Earthquake and tsunami that occurred on March 11, 2011, which triggered severe accidents at the Fukushima Dai-ichi nuclear power station (FDNPS). And the used filter tapes were sent to Tokyo Metropolitan University by the local governments through the Ministry of the Environment, Japan. The radionuclides in the hourly collected SPM samples at 99 of the more than 400 SPM stations were measured using Ge detectors. Because SPMs were collected hourly, the time series of the radiocesium concentration from March 12 to 23, 2011, has been described in detail. During this time, several radioactive plumes were observed in eastern Japan. Thus, precise description of these plumes (the peak time and the maximum radiocesium concentration in each plume at each SPM station) was our primary concern in this study. To confirm that our data were consistent with data independently measured by a research institution in Tokyo, we compared the hourly data by the institution with the hourly data for March 15-16 at an SPM station located near the institution. Although total suspended particulates were collected using a high-volume air sampler at the institution with a different sampling system, the data were highly consistent regarding the time series variations and radioactivity concentrations of ^{134}Cs and ^{137}Cs. This finding clearly indicates that the data presented in this study precisely reveal the ^{134}Cs and ^{137}Cs radioactivity concentrations in the atmosphere during March 2011. In addition to the radioactivity measurements, identifying the time and date when individual SPM samples were collected is equally important for producing a reliable database, and monthly reports of hourly SPM mass concentrations and SPM recording charts were used for confirmation. Cross-contamination is possible due to the apposition of radioactive materials from the SPM spot to the backside of the contacting tape. This cross-contamination is particularly likely for filter tapes made of polytetrafluoroethylene rather than glass fiber.

1. Introduction

Many estimates and evaluations have been made regarding internal dose exposure rates from inhalation,[1,2] atmospheric concentrations and/or the deposition densities simulated by atmospheric transport models[3,4] and the time-series release rate of radioactive materials just after the Fukushima Dai-ichi nuclear power station (FDNPS) accident.[5,6] In these studies, most of the data used were obtained from detailed deposition density maps of several radioactive nuclides in the surface soil, at local and regional scales by the surface or airborne measurements.[7,8] Those estimates and evaluations have large uncertainties because few data are available regarding the time-dependent concentrations of atmospheric radionuclides on a local/regional scale. In addition, datasets containing the deposition density of radionuclides provide no information regarding their time-series variations because they indicate only the values that accumulated by precipitation. Therefore, estimates of the internal radiation exposure from these data would be underestimated if atmospheric radionuclides passed through the area and were not precipitated.

In Japan, air quality is automatically monitored at many stations operated by the local governments throughout the Japanese islands under the air pollution control law. In this monitoring system, suspended particulate matter (SPM), which is defined as aerosols less than 10 μm in diameter, is collected on filter tape and its mass concentration is automatically measured using the β-ray attenuation method. Radioactive materials in atmospheric aerosols released by the FDNPS accidents could be collected on filter tapes in the SPM monitors, which were continuously operated even after the Great East Japan Earthquake and tsunami on March 11, 2011. By using the filter tapes installed in the SPM monitors at the air quality monitoring sites in the Fukushima Prefecture and the surrounding

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areas, we have developed a new methodology how to determine the spatio-temporal variation of radionuclides in atmospheric aerosols just after the accident, because there have been no reports of measurement methodologies for such variations. Eventually, we successfully measured radionuclides in SPM collected on filter tapes and published a paper that reported the spatio-temporal distribution of atmospheric ^{137}Cs in the Fukushima Prefecture and the Tokyo metropolitan area from March 12 to 23, 2011.[9]

The first aim of this paper is to summarize how to establish methods for measuring radionuclides in atmospheric aerosols collected on filter tapes installed in the SPM monitors in the air quality monitoring networks. The second aim is to present a dataset containing hourly atmospheric ^{134}Cs and ^{137}Cs concentrations from 99 SPM monitoring sites for users who are concerned with the transport of radioactive materials in the environment due to the FDNPS accident and their influences on human health.

A brief description of the history of this study performed for the first time is presented below. A test study regarding radionuclide measurements of used SPM filters was successfully conducted between February and April, 2012, in cooperation with the Ministry of Education, Culture, Sports, Science and Technology (MEXT) and the Ministry of the Environment (MOE), Japan. In the test study, atmospheric radionuclides in SPM samples at several sites were measured. A radioactivity of more than 1 Bq m^{-3} for ^{134}Cs and ^{137}Cs was determined when conducting one-hour measurements with Ge detectors. Unfortunately, ^{131}I could not be detected because of its short half-life of 8 days. After the test study, the SPM filter tapes used in March 2011 at more than 400 SPM stations were collected at Tokyo Metropolitan University (TMU) from many local governments in eastern Japan through the request of the Ministry of the Environment, Japan. Then, TMU and the Atmosphere and Ocean Research Institute (AORI) of the University of Tokyo began systematic measurements of radionuclides on the SPM filter tapes collected in the southern region of Tohoku including the Fukushima Prefecture and the Tokyo metropolitan area, for the periods of March 15 to 16 and March 20 to 23, 2011, when radioactive materials were widely transported in the major plumes and caused severe environmental pollution in eastern Honshu (the main island of Japan). The purpose of these measurements was to reveal when a radioactive plume reached each SPM station and to determine how high the maximum atmospheric radionuclide concentrations were at their peak in the plume. Most of the data obtained in FY 2012 are currently available to the public on the website.[10] Following the measurements in FY 2012, the systematic measurement of radionuclides of used SPM filter-tapes in many SPM stations was succeeded in FY 2013, when the SPM samples (mostly from the same but slightly expanded localities as mentioned above) were measured by Japan Chemical Analysis Center (JCAC) using the same methodology as that developed by TMU and AORI. The data obtained at JCAC are also open to the public on the website.[11]

The SPM sample measurements at TMU were carried out in FY 2012 and 2013 parallel to the measurements at JCAC and continued into FY 2014. These measurements included SPM filter samples that were collected at selected SPM sites during March 12 to 14 and March 17 to 19, 2011, when radioactive materials were transported only to the southern region of Tohoku.

In this paper, the data at 99 SPM stations from the local governments of Tokyo Metropolis, 8 prefectures (Yamagata, Miyagi, Fukushima, Ibaraki, Saitama, Chiba, Kanagawa, Niigata), and 13 cities (Sendai, Koriyama, Iwaki, Kawaguchi, Tokorozawa, Kawagoe, Chiba, Kashiwa, Hachioji, Yokohama, Yokosuka, Fujisawa, and Sagamihara) are summarized and presented in a unified format for wide use. The dataset includes the previously mentioned data that are available on the websites[10,11] and other data obtained by TMU and AORI which are not opened yet. Although the experimental methods and results presented in this study have been partially described elsewhere,[9,12] they are fully described in the following two sections.

2. Experimental methods

2.1. Collection of SPM samples. The monitoring sites of the used SPM samples analyzed in this study are listed in Table 1 with the longitude and latitude of each site. A code number is assigned for each monitoring station and is given in Table 1. In addition, the material of the filter tapes (either glass fiber (GF) or polytetrafluoroethylene (PTFE)), the flow rate used for SPM collection and the number of SPM samples collected on each roll of filter tape for one round are indicated. The locations of the SPM monitoring stations are shown in Figure 1 with their code numbers.

The filter tape rolls used in March 2011 at more than 400 SPM monitoring stations in eastern Japan were collected at TMU with the help of local governments and the MOE in 2012 and 2013. In this study, only portions of the SPM filter tapes were used for the radiocesium determinations. These tape rolls were usually stored in dark places by the individual local governments before being shipped to TMU.

An automatic SPM monitor is located at each air pollution monitoring station. This monitor vacuums up the ambient air through an inlet tube that is a few meters long at a flow rate of 15, 16.7 or 18 liters per minute, depending on the instrument model used. Atmospheric particulates with diameters of less than 10 μm are collected in the disk (11 mm or 16 mm in diameter) on a filter tape for one hour, and a mass concentration of SPM is automatically measured using the β-ray attenuation method. The SPM collected on the filter tape is usually visible as a dark disk, which is called an SPM spot in this article. The daily SPM collection begins at midnight (0:00) and ends 24 h later at midnight (24:00). Before beginning collection for the next day at 0:00, the tape is automatically wound forward by the same length for each spot, with no sampling to distinguish the change of collection date. This space with no SPM sample is called a blank spot in this article. As shown in Table 1, two types of materials (PTFE and GF) of the filter tapes were used depending on the model of the SPM monitoring instrument installed at each station. Among these materials, GF was more commonly used.

2.2. Identification of sampling date and time. Identification of the sampling date and time is the most important step in this study. Normally, a series of 24 SPM spots was collected for one day between two blank spots. Because each spot and/or each bunch of spots is not automatically marked, the sampling date and time must be identified for each spot manually by referring to the initial and/or final sampling date and time, which was normally marked on the edge of the tape or its container box by the local government when setting up and/or removing the tape. However, more than or fewer than 24 sampled spots were occasionally collected on the tape for one day. This occurred when an instrument was subjected to maintenance or encountered trouble in operation or a planned or unplanned electricity outage. In these cases, a monthly report of the hourly SPM mass concentrations released by the local government was helpful for identifying the sampling time. Because no mass concentration values are reported when the SPM collection temporarily stops, these missing values can be very informative for identifying the collection time. A chart paper was another important document for identifying the sampling date and time of the SPM spot. It usually draws an integrated mass concentration curve from zero to the maximum at

TABLE 1: Sampling sites of SPM and filter-tapes analyzed in this study (Both of Station name and Site are written in Japanese notation)

Code	Station name	Site	Longitude	Latitude	Tape[1]	Flow rate (L/min)	Number of spots per round[2]			
							March 12-14	March 15-16	March 17-19	March 20-23
			Miyagi Prefecture							
04-009	Shichigo	Wakabayashi-ku, Sendai-shi	140.94	38.24	GF	16.7	-	13	-	11
04-011	Yamada	Taihaku-ku, Sendai-shi	140.83	38.22	GF	18	10	16	-	18
04-023	Shiroishi	Shiroishi-shi	140.62	38.00	GF	18	-	16	-	18
04-025	Natori-jihai	Natori-shi	140.89	38.17	GF	18	-	18	-	20
04-028	Iwanuma	Iwamuma-shi	140.87	38.11	GF	15	-	14	-	16
04-029	Shibata	Shibata-machi	140.77	38.06	GF	15	-	20	-	21
04-030	Marumori	Marumori-machi	140.82	37.86	GF	15	-	-	16	17
04-032	Matsushima	Matsushima-machi	141.07	38.39	GF	18	-	20	=	=
04-036	Kokusetsu-nonodake	Wakutani-machi	141.17	38.55	PTFE	18	-	=	=	=
04-037	Tsukidate	Kurihara-shi	141.02	38.74	GF	18	-	18	-	19
			Yamagata Prefecture							
06-001	Yamagata-tokamachi	Yamgata-shi	140.34	38.25	GF	18	-	16	-	18
06-005	Yonezawa-kanaike	Yonezawa-shi	140.11	37.92	GF	18	-	17	-	18
			Fukushima Prefecture							
07-001	Furukawa	Fukushima-shi	140.49	37.78	GF	16.7	=	=	=	11
07-002	Minamimachi	Fukushima-shi	140.46	37.74	PTFE	18	-	14	-	14
07-003	Moriai	Fukushima-shi	140.45	37.77	GF	16.7	-	7	-	11
07-004	Sugitsumacho	Fukushima-shi	140.47	37.75	GF	18	-	19	-	21
07-005	Aizuwakamatsu	Aizuwakamatsu-shi	139.92	37.49	GF	15	-	22	-	23
07-006	Asahi	Koriyama-shi	140.36	37.41	GF	18	-	18	=	20
07-012	Daishin	Koriyama-shi	140.34	37.39	GF	18	-	17	-	19
07-019	Atagoshita	Iwaki-shi	140.9	36.95	GF	18	-	-	-	15
07-020	Ohara	Iwaki-shi	140.89	36.96	GF	18	-	-	-	12
07-030	Shirakawa	Shirakawa-shi	140.22	37.12	GF	16.7	-	11	-	13
07-031	Haramachi	Minamisoma-shi	140.95	37.64	GF	16.7	-	12	-	11
07-032	Sukagawa	Sukagawa-shi	140.37	37.29	GF	16.7	-	12	-	13
07-033	Kitakata	Kitakata-shi	139.87	37.66	GF	18	-	18	-	20
07-034	Soma	Soma-shi	140.92	37.80	PTFE	18	-	14	15	12
07-035	Nihonmatsu	Nihonmatsu-shi	140.46	37.59	GF	18	-	17	-	19
07-036	Minami-aizu	Minamiaizu-machi	139.78	37.20	GF	15	-	25	-	24
07-037	Yabuki	Yabuki-machi	140.34	37.20	GF	18	-	23	-	25
07-038	Tanakura	Tanakura-machi	140.38	37.02	GF	18	-	=	-	25
07-043	Shinchi	Shinchi-machi	140.91	37.87	GF	18	-	16	-	=
			Ibaraki Prefecture							
08-009	Tsuchiura hokenjyo	Tsuchiura-shi	140.19	36.07	GF	18	-	17	-	19
08-010	Tsuchiura-nakamura-minami	Tsuchiura-shi	140.17	36.04	GF	18	-	19	-	21
08-011	Koga hokenjyo	Koga-shi	139.72	36.20	GF	18	-	13	-	16
08-015	Shimotsuma	Shimotsuma-shi	139.96	36.18	GF	18	-	16	-	18
08-022	Toride shiyakusyo	Toride-shi	140.05	35.91	GF	18	-	15	-	16
08-023	Tsukuba-kouya	Tsukuba-shi	140.02	36.10	GF	18	-	15	-	17
08-032	Kamisu-shimohataki	Kamisu-shi	140.63	35.92	PTFE	16.7	-	12	-	12
08-036	Kamisu-yokose	Kamisu-shi	140.7	35.85	PTFE	16.7	-	=	-	12
08-041	Moriya	Moriya-shi	139.98	35.95	GF	16.7	-	23	-	24
			Saitama Prefecture							
11-005	Kawagoeshi-kawagoe	Kawagoe-shi	139.49	35.93	GF	18	-	21	-	15
11-009	Kumagaya-koiduka-jihai	Kumagaya-shi	139.39	36.16	GF	18	-	20	-	22
11-026	Tokorozawashi-higashi-tokorozawa	Tokorozawa-shi	139.52	35.80	GF	18	-	22	-	17
11-037	Kasukabe	Kasukabe-shi	139.75	35.97	GF	18	-	16	-	17
11-041	Konosu	Konosu-shi	139.52	36.07	GF	18	-	21	-	22/14[a]
11-042	Konosu-tenjin-jihai	Konosu-shi	139.52	36.06	GF	18	-	19	-	20
11-043	Fukaya	Fukaya-shi	139.28	36.19	GF	18	-	=	-	=
11-057	Wako	Wako-shi	139.62	35.78	GF	18	-	20	-	21
11-060	Kuki	Kuki-shi	139.66	36.06	GF	18	-	17	-	18
11-064	Misato	Misato-shi	139.88	35.84	GF	18	-	19	-	21
11-071	Ogawa	Ogawa-machi	139.26	36.06	GF	18	-	20	-	22
11-072	Higashi-chichibu	Higashichichibu-mura	139.19	36.01	GF	18	-	17	-	19
11-073	Honjyo-kodama	Honjyo-shi	139.13	36.19	GF	18	-	20	-	21
11-075	Yorii	Yorii-machi	139.19	36.12	GF	18	-	17	-	18
11-077	Kankyokagaku Kokusai senta	Kazo-shi	139.56	36.08	GF	18	-	21	-	-
11-080	Kawaguchishi-shiba	Kawaguchi-shi	139.69	35.83	GF	18	-	15	-	17

TABLE 1: Continued

Code	Station name	Site	Longitude	Latitude	Tape[1]	Flow rate (L/min)	Number of spots per round[2]			
							March 12-14	March 15-16	March 17-19	March 20-23
			Chiba Prefecture							
12-004	Sanno shogakkou	Inage-ku, Chiba-shi	140.14	35.66	GF	18	-	=	-	22/15[b]
12-009	Chishirodai-kita shogakkou	Wakaba-ku, Chiba-shi	140.18	35.63	GF	18	-	18	-	20
12-019	Toke	Midori-ku, Chiba-shi	140.26	35.53	GF	16.7	-	13	-	15
12-047	Tateyama-kamegahara	Tateyama-shi	139.88	35.02	PTFE	18	-	14	-	14
12-048	Izumiya shogakkou	Midori-ku, Chiba-shi	140.17	35.55	GF	18	-	19	-	=
12-049	Kisarazu-chuo	Kisarazu-shi	139.93	35.38	GF	18	-	16	-	18
12-057	Nodashi-noda	Noda-shi	139.87	35.95	GF	18	-	16	-	17
12-060	Mobara-takashi	Mobara-shi	140.29	35.43	GF	18	-	17	-	18
12-061	Narita-karabe	Narita-shi	140.30	35.77	GF	18	-	17	-	18
12-065	Togane-horiage	Togane-shi	140.37	35.56	PTFE	18	-	13	-	=
12-070	Kashiwa-omuro	Kashiwa-shi	139.96	35.90	GF	18	-	18	-	20
12-073	Katsuura-kobado	Katsuura-shi	140.27	35.18	PTFE	18	-	16	-	16/13[c]
12-093	Abiko-kohokudai	Abiko-shi	140.08	35.86	GF	15	-	16	-	17
12-094	Kamagaya-karuizawa	Kamagaya-shi	140.02	35.79	GF	15	-	19	-	22
12-096	Kimitsu-kubo	Kimitsu-shi	139.90	35.33	GF	18	-	16	-	17
12-116	Inzai-takabana	Inzai-shi	140.13	35.79	PTFE	18	-	16	-	16
12-117	Sakae-ajikidai	Sakae-machi	140.25	35.85	PTFE	18	-	16	-	16
12-118	Narita-nado	Narita-shi	140.42	35.85	PTFE	18	-	16	-	14
12-120	Katori-fuma	Katori-shi	140.61	35.79	PTFE	18	-	14	-	14
12-123	Ichinomiya-torami	Ichinomiya-machi	140.38	35.35	PTFE	18	-	14	-	=
12-134	Sodegaura-ozone	Sodegaura-shi	140.02	35.42	GF	15	-	13	-	14
			Tokyo Metropolitan Prefecture							
13-023	Himonya	Meguro-ku	139.68	35.62	PTFE	18	-	14	-	=
13-025	Kannanadori-kakinoki zaka	Meguro-ku	139.68	35.62	GF	15	-	15	-	17
13-027	Kannanadori-matsubara bashi	Ota-ku	139.71	35.60	PTFE	18	-	14	-	15
13-028	Nakaharakaido-minami senzoku	Ota-ku	139.69	35.60	PTFE	18	-	14	-	14
13-031	Setagayaku-seijyo	Setagaya-ku	139.59	35.65	PTFE	18	-	14	-	15
13-052	Katsushikaku-mizumoto-koen	Katsushika-ku	139.87	35.79	PTFE	18	-	13	-	14
13-056	Edogawaku-minamikasai	Edogawa-ku	139.87	35.65	PTFE	18	-	13	-	14
13-058	Hachiojishi-tatemachi	Hachioji-shi	139.29	35.63	GF	18	-	15	-	16
13-065	Omeshi-higashiome	Ome-shi	139.28	35.79	PTFE	18	-	=	-	15
13-069	Machidashi-nogaya	Machida-shi	139.48	35.59	PTFE	18	-	15	-	16
13-089	Hachiojishi-Kawaguchimachi	Hachioji-shi	139.28	35.70	GF	18	-	22/15d	-	16
			Kanagawa Prefecture							
14-008	Nakaku-honmoku	Naka-ku, Yokohama-shi	139.66	35.41	PTFE	18	-	13/15[e]	-	13
14-050	Kurihama gyoseisenta	Yokosuka-shi	139.71	35.23	GF	18	-	16	-	17
14-060	Shonandai bunkasenta	Fujisawa-shi	139.47	35.40	GF	18	-	22	-	16
14-064	Odawara shiyakusyo	Odawara-shi	139.15	35.26	GF	18	-	-	-	13
14-065	Odawara shiminkaikan	Odawara-shi	139.16	35.25	PTFE	18	-	14	-	14
14-074	Tsukui	Midori-ku, Sagamihara-shi	139.26	35.59	PTFE	18	-	15	-	-
14-077	Misaki chugakkou	Miura-shi	139.62	35.14	GF	18	-	23	-	24
14-078	Hadano shiyakusho	Hadano-shi	139.22	35.37	PTFE	16.7	-	7	-	7
14-079	Hadanoshi-honcho	Hadano-shi	139.23	35.37	PTFE	16.7	-	12	-	12
14-085	Isehara shiyakusho	Isehara-shi	139.32	35.40	PTFE	16.7	-	12	-	12
			Niigata Prefecture							
15-030	Muikamachi	Minamiuonuma-shi	138.87	37.07	GF	18	-	11	-	14

1. Filter tape materials (GF: glass fiber; PTFE: polytetrafluoroethylene).
2. -: not counted, =: missing. Two numbers separated by a slash indicate that the filter-tape rolls were changed. The change was performed at the following times and dates; a: 12:00 on March 23, b: 12:00 on March 23, c: 9:00 on March 22, d: 11:00 on March 15, e: 13:00 on March 15.

the end of the measurement for each hour. In addition, the original SPM concentration for each hour is recorded on a chart with the collection time in hours. Furthermore, if routine maintenance is performed or a sudden blackout occurs, the stopping and starting time of the electricity is printed. Another advantage of using these documents was that, when a plume with high radioactive materials in SPM passed SPM monitoring stations, irregular/sudden decrease to a very low/zero value for SPM mass concentrations was frequently observed due to strong β-ray emission. By referring to these supporting materials, the identification and validation of the sampling date and time could be secured for almost all SPM spots in the database.

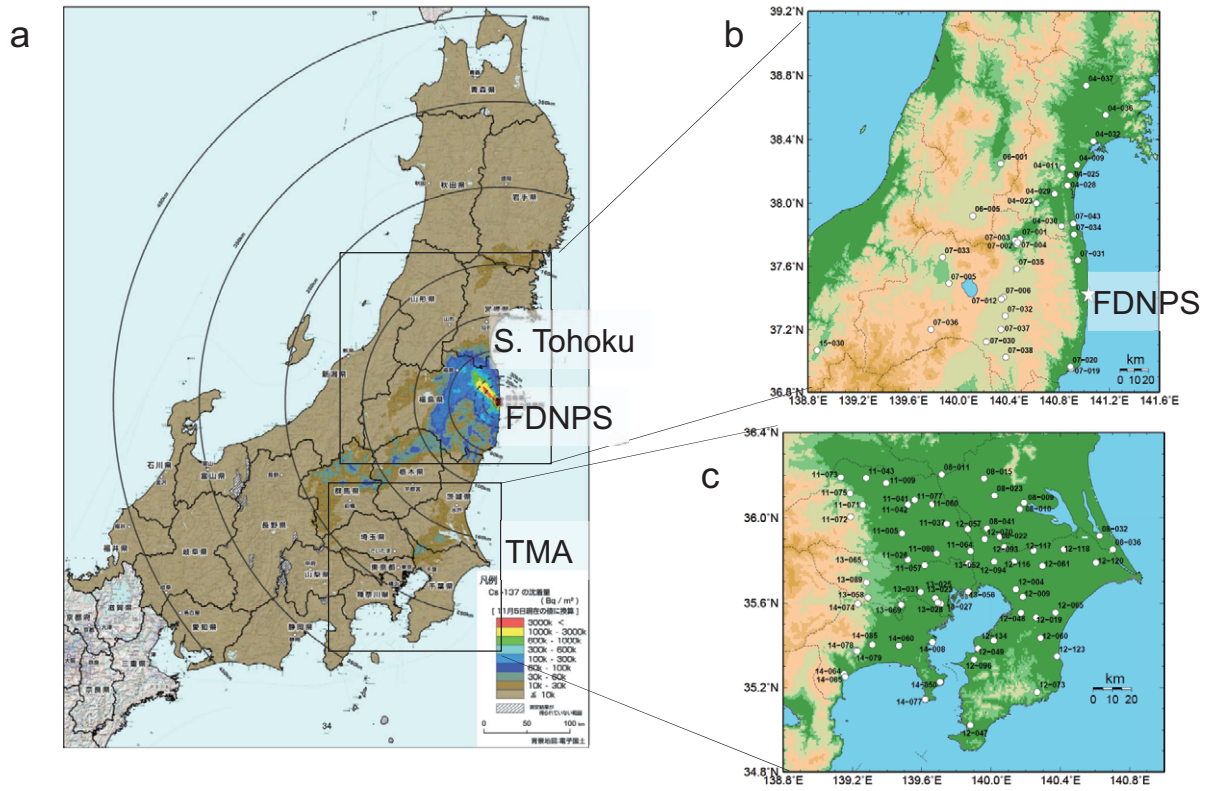

Figure 1. Sampling sites for the SPM samples in the air pollution monitoring network that were analyzed for ^{134}Cs and ^{137}Cs radioactivity concentrations. Two areas, i.e., S. Tohoku (southern Tohoku) and TMA (Tokyo metropolitan area), shown in Figure 1a, are expanded in Figures 1b and 1c, respectively. The deposition density of ^{137}Cs on the ground surface in eastern Japan from the Fourth Airborne Monitoring Survey by MEXT[8] is shown in Figure 1a for information. The code numbers are indicated in Figures 1b and 1c for reference.

2.3. Gamma-ray spectrometry. The filter tape was cut in the center between every two successive spots. A rectangular piece of tape including an SPM spot was sandwiched between weighing papers and was fixed on a thin plastic sheet by adhesive tape before subjecting to gamma-ray measurements. The SPM samples were prepared so that they could be reused for other purposes after the gamma-ray measurements. The gamma-ray measurements were performed at four different facilities, TMU, the Nuclear Professional School of the University of Tokyo (UT-NPS), the Japan Atomic Energy Agency (JAEA), and JCAC. Each sample that included a blank spot was measured for 1 to 3 hours using a Ge semiconductor detector. Several blank samples were measured for approximately 12 hours to monitor any possibility of contamination in the SPM samples. An example of gamma- ray spectrum of an SPM sample was shown in Figure 2.

The counting efficiencies of the Ge detectors for gamma-rays from ^{134}Cs and ^{137}Cs were determined by using homemade standard source samples of the same size as the SPM spot samples, which included known ^{134}Cs and ^{137}Cs activities. Two sets of these standard samples were independently prepared by TMU and JCAC. The typical detection limit was between approximately 0.1 and 0.3 Bq m^{-3} for the one-hour measurement when using most of the detectors. However, one detector had a higher detection limit of 1 to 2 Bq m^{-3}.

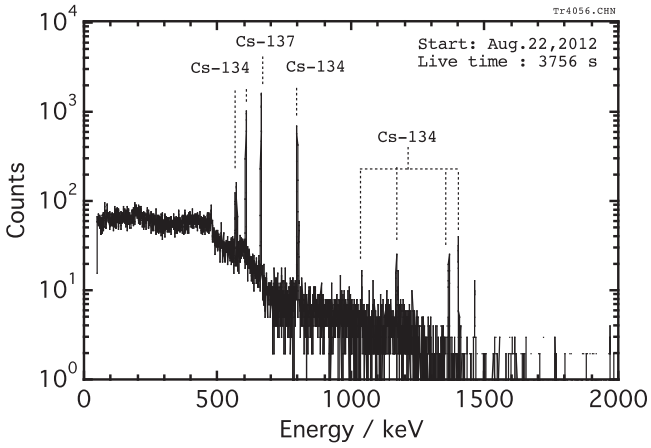

Figure 2. Gamma-ray spectrum of an SPM sampled at Kakinokizaka station (station code# 13-025) in Tokyo from 10:00 to 11:00 on March 15, 2011. The SPM sample was fixed at a distance of 4 mm from a Ge detector (GMX-30200S, ORTEC). A net area of 662 keV gamma-ray from ^{137}Cs counted 6973, corresponding to 50 Bq.

3. Results and Discussions

3.1. Data validation

3.1.1. Consistency in the activity data obtained at four different facilities. As described in section 2.3, the standard source samples of ^{134}Cs and ^{137}Cs were prepared separately at TMU and JCAC. These samples were used first or second to quantify the radioactivity of ^{134}Cs and ^{137}Cs in each SPM sample at the four facilities (TMU, UT-NPS, JAEA and JCAC). Thus, an inter-comparison of gamma-ray measurements was conducted between TMU and JCAC. Figure 3 compares the radioactivity values of ^{134}Cs and ^{137}Cs for the same set of 37 SPM spot samples determined at TMU and JCAC. Each facility used its own standard source sample and Ge detectors. The ^{134}Cs and ^{137}Cs radioactivity in the 37 SPM samples spreads from around the determination limit to 180 Bq m^{-3}. As shown in Figure 3, excellent agreement was confirmed between TMU and JCAC when determining the activity concentration range of 1 to 180 Bq m^{-3}. Such consistency guarantees the high reliability of the preparation of standard source samples and the gamma-ray spectrometry procedure at both facilities. For

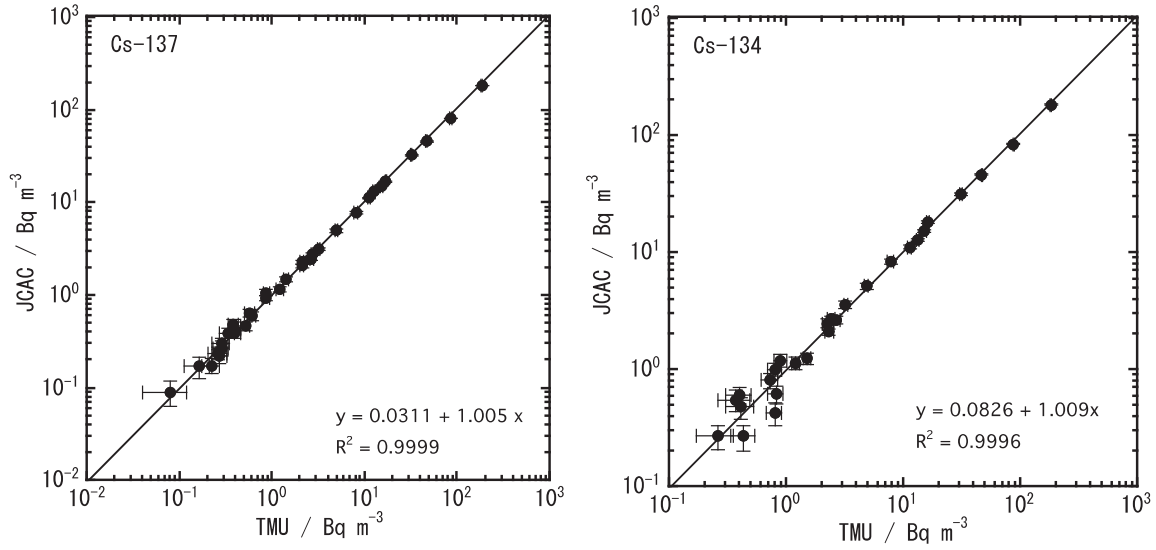

Figure 3. Comparison of ^{134}Cs and ^{137}Cs radioactivity concentrations (in Bq m^{-3}) determined at TMU and JCAC for the same SPM samples. The lines in the graphs show 1:1 relationships between TMU and JCAC rather than regression lines of measured values.

TABLE 2: Comparison of the sampling systems of atmospheric aerosols between the SPM monitoring stations and TIRI

Items	SPM monitors	Equipment at TIRI
Inlet tube	a few meters long	direct sampling with no tube
Diameter of sampled aerosols	< 10 μm	all (usually < 30 μm)
Diameter of sampled spot circle	11 mm or 16 mm	100 mm
Material of filters*	GF or PTFE	GF
Airflow rate (L m^{-3})	15-18	600
Sampling Interval (hour)	1	1-3, 8, 24

* GF: glass filter, PTFE: polytetrafluoroethylene.

^{134}Cs, data points smaller than 1 Bq m^{-3} are scattered approximately 1:1, but the values of TMU and JCAC are consistent within 2σ error uncertainty. Thus, it can be concluded that the data quality obtained at TMU and JCAC is equally high.

Calibration of the ^{134}Cs and ^{137}Cs radioactivity at UT-NPS and JAEA, where ^{134}Cs and ^{137}Cs standard source samples were not used, was performed using some SPM samples with ^{134}Cs and ^{137}Cs activity concentrations that were determined at TMU by using their own standard source samples. Although the uncertainty of analytical results obtained at UT-NPS and JAEA should have increased owing to the propagation of errors, this increment was very small because the SPM samples used for calibration were measured long enough so that the errors due to counting statistics became negligibly small.

3.1.2. Consistency in the activity data obtained by different monitoring systems. In Japan, SPM samples have been collected at air pollution monitoring stations for several decades. According to the Air Quality Standards set in Japan in 1973, SPM is defined as suspended aerosols with diameters of less than 10 μm. Most monitoring stations continued to operate after the Great East Japan Earthquake and tsunami. Meanwhile, several institutions began to independently measure radionuclides in the atmosphere soon after the FDNPS accident in March 2011. Among them, the Tokyo Metropolitan Industrial Technology Research Institute (TIRI) began measuring radionuclides, including ^{137}Cs, in the total suspended particulates (TSPs) at Fukasawa, Setagaya-ku, Tokyo,[13] so that aerosols even larger than 10 μm in diameter were also collected. TIRI directly collected TSPs on glass fiber filters (100 mm ϕ) with no inlet tubes using a high-volume air sampler at a flow rate of 600 L min^{-1} for 1-3, 8, or 24 hours, depending on the gamma-ray dose rate at the sampling site. The gamma-rays of the TSPs were measured using a Ge detector for 1,000 to 20,000 seconds and the radionuclide concentrations were promptly presented on the website. There are more than 50 SPM monitoring stations in the Tokyo Metropolis. Among them, the Kakinokizaka station (code #: 13-025) is located approximately 1.6 km away from the TIRI sampling site, where an SPM sampler using glass fiber filter tape was in operation in March 2011. The sampling systems of atmospheric aerosols used at the SPM monitoring stations and at TIRI are compared in Table 2.

Figure 4 shows the time series of ^{137}Cs concentrations in aerosols on the filters at Fukasawa[13] and Kakinokizaka (from this work). As shown in Table 2, the SPM collection system at Kakinokizaka was significantly different from that at Fukasawa. Furthermore, the SPM samples collected at Kakinokizaka were measured more than one year after collection. Nevertheless, excellent consistency was observed between the two independent measurements. The time of the peak and its corresponding activity concentrations are nearly the same. This finding clearly indicates that the SPM filter tape quantitatively preserves atmospheric radiocesium-containing materials for more than a year, and that the SPM monitor could collect nearly all the atmospheric radiocesium in the particulate phase just after the accident. In addition, this result suggests that the time series variations of these radioactive nuclides in the atmosphere can be reproduced. Hence, hourly ^{134}Cs and ^{137}Cs activity values in the SPM samples can be used in further scientific studies as reliable data for radiocesium concentrations in the atmosphere.[9,12]

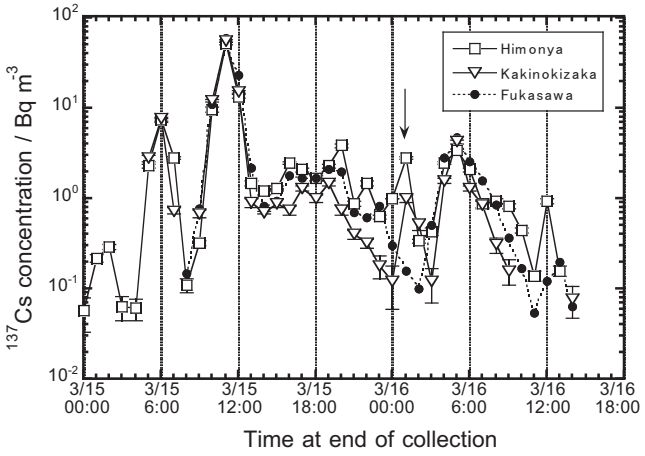

Figure 4. Time series variation of atmospheric ^{137}Cs concentrations at three sites in Tokyo between March 15 and 16. The SPM samples analyzed in this study were collected at the Himonya and Kakinokizaka SPM monitoring stations which are shown in Figure 1c, and the samples from Fukasawa were collected by the Tokyo Metropolitan Industrial Technology Research Institute (TIRI). These three sites are located near each other. The concentrations below the detection limit at Kakinokizaka (approximately 0.1 Bq m^{-3}) are not shown. The data shown by an arrow for Himonya and Kakinokizaka SPM stations are assumed false (see the text). At Fukasawa, the SPM sampling durations used for the first three data points on the morning of March 15 were 0:00-7:12, 7:12-8:23, and 8:23-9:00 (JST). From the next sample, a sampling time of exactly one hour was fixed. The averaged ^{137}Cs concentration in the first SPM sample was 1.8 Bq m^{-3}, according to Reference 13 (data not shown).

3.2. Cross-contamination. Although excellent agreement can be confirmed in the time series variations of the ^{137}Cs activity concentrations between the two datasets (SPM and TSPs) in Figure 4, an inconsistency can be noted. A small peak appears at 1:00 on March 16 in the Kakinokizaka data, while the ^{137}Cs concentration by TIRI consistently decreased with no such peak. In Figure 4, the ^{137}Cs concentration variations of the SPM samples collected at the Himonya station (code #: 13-023) are also shown, which is located approximately 500 m south-southeast of the Kakinokizaka station (13-025). The concentration variations of ^{137}Cs at these two SPM monitoring stations are generally consistent with each other and with those determined by TIRI. However, the small peak at 1:00 on March 16 is considerably enhanced in the Himonya data compared with that of the Kakinokizaka data (an arrow in Figure 4). Because this peak was not observed at 1:00 at Fukasawa, the peak was thought to be a ghost peak that resulted from mechanical factors during the SPM filter tape sampling. When the data and the filter tape were carefully inspected, cross-contamination was suspected to explain this discrepancy.[9,12] Because the filter tape was tightly wound, portions of the aerosols that included radioactive materials in the SPM samples were potentially transferred to the backside of the contacting tape. At this sampling time, we confirmed that 14 and 15 SPM spots were included in one round of the tapes used at Himonya and Kakinokizaka, respectively. As a result, the peak top spot at 11:00 on March 15, when the maximum concentrations of 52.3 and 55.6 Bq m^{-3} were measured at Himonya and Kakinokizaka, respectively, faced the backside of a blank spot between March 15-16 and the next SPM spot (collected at 1:00 on March 16) on the contacting tape just one round after. In fact, a high ^{137}Cs activity concentration (8.3 Bq m^{-3}, assuming that the same volume of air for an SPM spot sample was vacuumed) was measured for the blank spot of Himonya, whereas that of Kakinokizaka was very low (0.08 Bq m^{-3}). The high concentration value at Himonya was lowered to 1.6 Bq m^{-3} by wiping the backside of the blank spot tape and could not be further lowered by additional wiping. Based on these considerations and measurements, the peaks that appeared at 1:00 on March 16 were erroneous and were caused by the transfer of a portion of the radioactive materials in the SPM collected at 11:00 on March 15 onto the backside of the contacting tape. Contributions to the backside were estimated to be 15% for Himonya based on the geometry of the spot arrangement and the ^{137}Cs activity in the blank spot.

In this study, the contribution of cross-contamination (Cc in %) is defined as Cc=(Cb/(Cp+Cb))×100, where Cp and Cb (in Bq m^{-3}) are the highest concentrations at the peak time, and the concentration of the sampled or blank spot corresponding with one round after the peak spot, respectively. For the Kakinokizaka station, the contribution of cross-contamination was estimated to be 1.8%, because the highest concentration (Cp) occurring at a peak time of 11:00 on March 15 and the concentration (Cb) of the spot sampled at 1:00 on March 16, which corresponded to the 15th spot from the peak spot for one round, were 55.6 and 1.0 Bq m^{-3}, respectively. Here, it was assumed that all the ^{137}Cs radionuclides in the spot sampled at 1:00 on March 16 were contributed by the sample at 11:00 on March 15. At Himonya, if all of the ^{137}Cs in the blank spot (8.3 Bq m^{-3}, corresponding to Cb) were transferred from the spot with the highest concentration (Cp, 52.3 Bq m^{-3}) of ^{137}Cs at a peak time of 11:00, the contribution (Cc) was 14%, which is nearly equal to the contribution of 15% provided by the above estimation.

The difference in the level of cross-contamination observed at the two sampling stations (Himonya and Kakinokizaka) can be explained by the difference in the materials used for the filter tape. The filter tape used at Himonya was made of PTFE, while that used at Kakinokizaka was glass fiber (GF). As reported in the previous paper,[9] there are several reasons why the radioactive materials adhering to the PTFE filter tape are transferred to the contacting tape more easily than those adhering to the GF tape; (i) the thickness of the PTFE filter is approximately 30% of that of the GF filter, which allows the PTFE tape to be tightly wound, (ii) SPM can penetrate inside of the GF filter more deeply than the PTFE filter because of the coarse structure of the GF, and (iii) the PTFE filter is easily charged with static electricity. It should be noted that the transfer of radioactive materials underestimates and overestimates the activity concentration of SPM samples around the peak time in a plume and at the late time after one round, respectively.

We carefully investigated the contributions of cross-contamination (Cc) using the individually measured ^{137}Cs concentrations at the SPM stations where the PTFE filter tape was used. At this stage, the Cc was estimated to be less than 10% when the ^{137}Cs concentration was higher than 70 Bq m^{-3} around the peak time, except for one data that resulted in a Cc of 19% (at 10:00 on March 16 at the station of 12-120). By contrast, the Cc was less than 15% when the ^{137}Cs concentration was between 20 and 70 Bq m^{-3}. The reason why the contributions for the lower peak concentrations (20-70 Bq m^{-3}) were higher than those for the higher peak concentrations (> 70 Bq m^{-3}) has not been clarified until now. Hence, the contribution of Cc was independent of the magnitude of the high ^{137}Cs concentration (> 20 Bq m^{-3}) around the peak time. In fact, although the peak ^{137}Cs concentrations at the two SPM stations were nearly equal, the contributions were different from each other. On the contrary, the Cc for the SPM stations that used a GF filter tape was less than 3% and 5% when the peak ^{137}Cs concentration was greater than 100 Bq m^{-3} and between 20 and 100 Bq m^{-3}, respectively. These results clearly indicate that the Cc for the SPM stations using GF was much smaller than that for the stations using PTFE.

Cross-contamination due to the transfer of radioactive materials was observed not only at the Himonya and Kakinokizaka stations but also at other monitoring stations. Such cross-con-

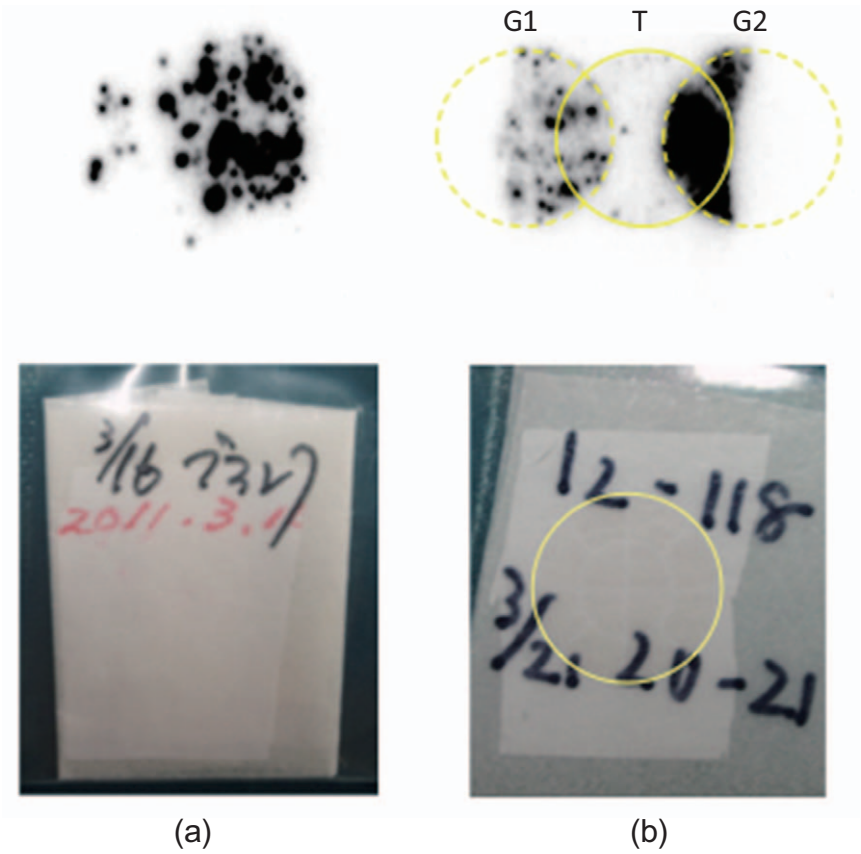

Figure 5. Typical autoradiographs (upper figure) obtained by using an imaging plate and photographs (bottom figure) of SPM samples. (a) A blank sample between March 15 and 16 at the Himonya station (code #: 13-023, PTFE filter). Although there was not any visible SPM on the blank filter, some radioactive materials were detected by autoradiography. (b) An SPM sample collected from 20:00 to 21:00 on March 21 at the Narita-Nado station (#12-118, PTFE filter). Although only one circular SPM was faintly visible in the photograph, one circular and two semicircular images were observed in the autoradiograph. Yellow circles are artificially drawn for better understanding. An image edged with circle T is a radiograph of actually sampled spot, and images edged with circles G1 and G2 are radiographs of ghost spots attached on the backside of the filter tape. Both of G1 and G2 are estimated to be portions of SPM sampled at 8:00 and 7:00 on March 21, respectively, when a plume passed the station, considering that the total spot number was 14 per one round, as shown in Table 1.

tamination could be easily recognized by autoradiography imaging with an imaging plate. Examples of these images and photographs of the samples are shown in Figure 5. Figure 5 (a) shows an autoradiograph of a blank sample between March 15 and 16 at the Himonya station (code#: 13-023) described above, which should not have contained any SPM as shown in the photograph. However, an approximate circle image and an image of a small piece in the left side are clearly visible, which imply that some radioactive materials were present on the blank spot tape and that these materials were transferred to the back surface of the tape from the contacting spot having large amounts of radioactive materials from a plume. Another case is shown in Figure 5 (b), in which three images can be noticed, i.e., one faint complete circle disk (T) and two clear incomplete broken disks (G1 and G2). The complete disk is a real SPM spot collected between 20:00 and 21:00 on March 21 at the Narita-Nado station (code #: 12-118) with the use of PTFE filter tapes. Two incomplete discs were transferred, probably from the spot at 6:00-7:00 and 7:00-8:00 on the same day when high ^{137}Cs activity concentrations of 148 and 57.2 Bq m^{-3} were measured due to a plume, respectively. Even when all the radionuclides of ^{137}Cs concentrations (11.4 Bq m^{-3}) in the sampled spot at 21:00 were supposed to be attached to the backside from the surface of the spot sampled at 7:00, the contribution to the backside was only 7.2%, which was much lower than that (14-15%) estimated for the Himonya station. From the IP intensity, approximately 90% of the ^{137}Cs activity at 21:00 was estimated to be contributed from the transferred SPM. Thus, the real ^{137}Cs activity of this spot cannot be accurately obtained. We are now preparing a technical report that describes these important results in detail.[14]

In this paper, no additional information on the effect of cross contamination as described for the Himonya and Narita-Nado stations is shown in the tables of Appendix A, because we have not finished checking all the suspicious SPM samples by autoradiography. We have been judging one by one whether the ^{137}Cs concentrations of a few sampled spots located one round after the previously sampled spots with the highest ^{137}Cs concentrations in a plume, are higher due to cross contamination than actual ^{137}Cs concentrations which are estimated to be much lower. After completing this assessment, we will soon publish a technical report and revise the dataset reported in this paper, showing all the data which should not be used for further analysis due to overestimation.

3.3. Analytical data presentation. The numerical data of the activity concentrations (in Bq m^{-3}) are tabulated in Appendix A, Tables A1 through A99. Radioactivity values are decay-corrected to the time when the collection of SPM samples ended. Each table corresponds to each sampling site. No information for cross-contamination were made. The detection limit is defined as 3σ of the total background counts (C_{bk}) at the peak areas around 605 keV and 662 keV for ^{134}Cs and ^{137}Cs, respectively, in gamma-ray spectrometry, where σ is a square root value of C_{bk}. Examples of time series variations of the radioactive ^{137}Cs concentrations (in Bq m^{-3}) discussed in Section 3.2 are shown in Figure 6a to 6c. Time series variations of ^{137}Cs concentrations at all SPM stations are shown in

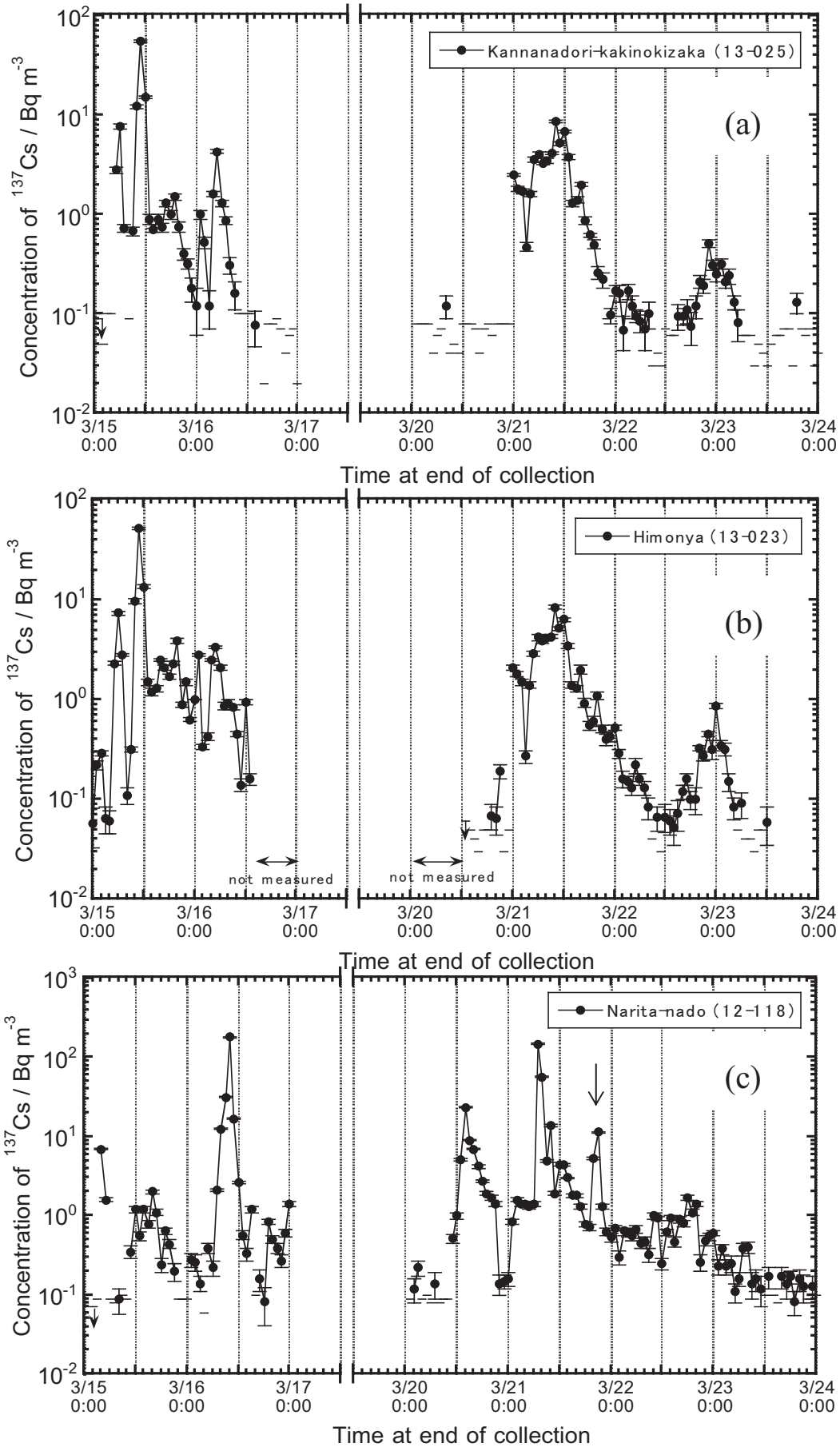

Figure 6. Time series variation of atmospheric ^{137}Cs concentration observed at (a) Kannanadori-kakinokizaka station (13-025) in Meguro-ku, Tokyo (also Figure B75), (b) Himonya station (13-023) in Meguro-ku, Tokyo (also Figure B78), (c) Narita-nado station (12-118) in Narita-shi, Chiba (also Figure B74), (d) Moriai station (07-003) in Fukushima-shi, Fukushima (also Figure B15) and (e) Minamimachi station (07-002) in Fukushima-shi, Fukushima (also Figure B14). The data shown by an arrow for the Narita-nado station (c) is assumed false (see Section 3.2 and Figure 5 of the text).

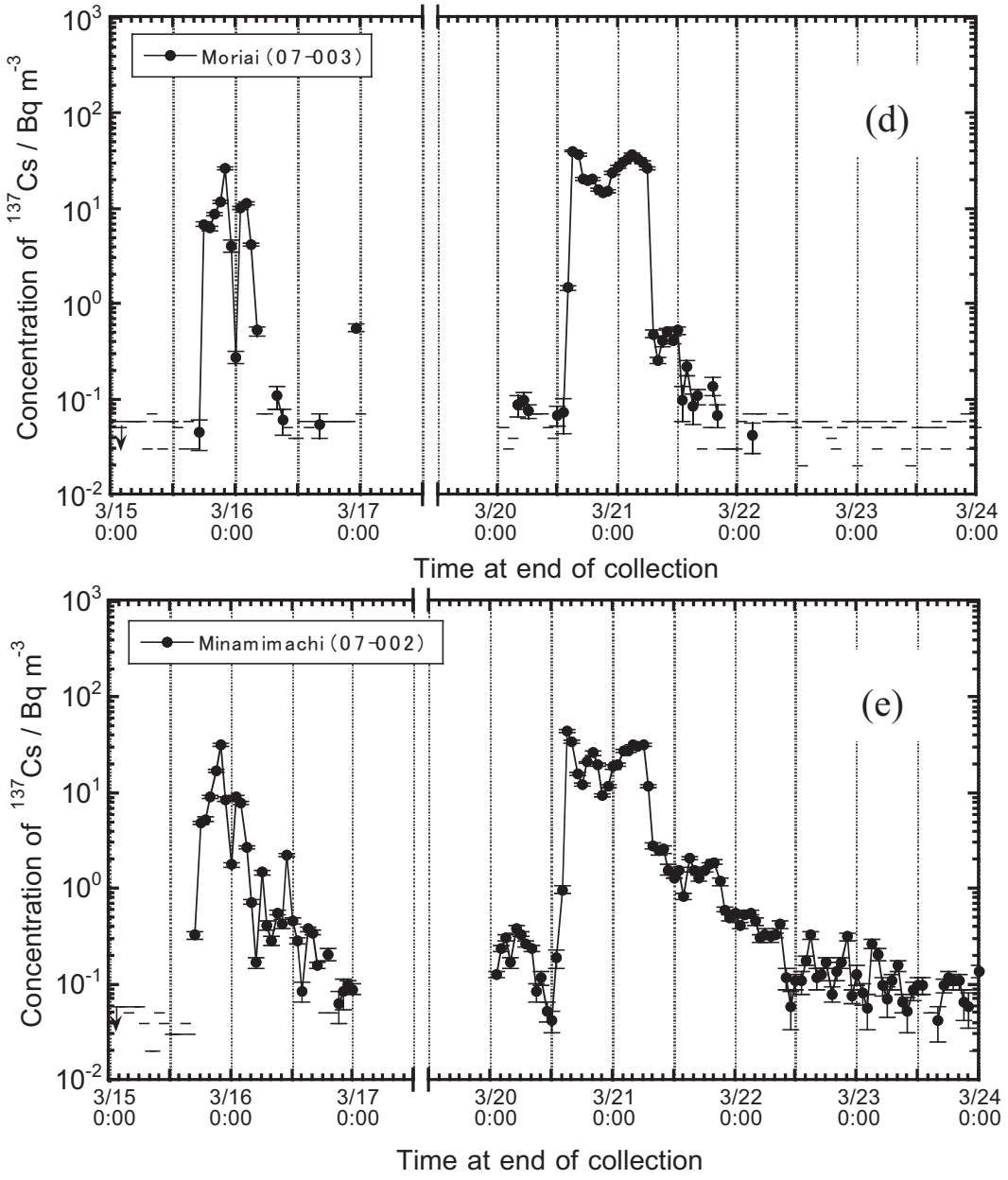

Figure 6. Continued.

Appendix B, Figure B1 to B99. Although ^{134}Cs activity was measured along with that of ^{137}Cs, its variations are not shown because its time series variations are essentially the same as those for ^{137}Cs, as implied by the nearly constant activity ratios (approximately 1) of ^{134}Cs/^{137}Cs for most SPM samples analyzed. Some SPM samples collected at Haramachi (code #: 07-031), Soma (07-034) and Shinchi (07-043) stations on March 12, 2011 had obviously smaller ratios of approximately 0.9.[15] The precise determination of ^{134}Cs/^{137}Cs activity ratios will be reported elsewhere.

3.4. Some implications. In our first paper[9] we presented the hourly spatial distribution of ^{137}Cs in the Fukushima Prefecture and the Tokyo metropolitan area, which is located more than 170 km southwest of FDNPS, just after the accident by using hourly radiocesium concentration data from the SPM filter samples collected at the 40 SPM stations from March 12 to 23, 2011. In the paper, we first revealed how the radioactive materials released into the atmosphere following the accident in March 2011 were transported in a local/regional scale. The content of our study in the paper is summarized below.

1. Nine major plumes with ^{137}Cs concentrations greater than 10 Bq m^{-3} were found between March 12 and 23. Of these, five and four plumes were transported to the Fukushima Prefecture and the Tokyo metropolitan area, respectively. The transport of radioactive materials from the FDNPS to the northern part of Hamadori in the east coast area of the Fukushima Prefecture, which was observed four times, could be firstly recognized by measuring radiocesium in the SPM samples.
2. Two plumes transported to the Tokyo metropolitan area were observed on March 16 and 20, 2011 for the first time, in addition to the two previously recognized major plumes on March 15 and 21, 2011.
3. The radiation dose rate measured at the monitoring posts in Nakadori in the central part of the Fukushima Prefecture did not increase, even when the plume passed by on March 20, 2011, because the dose rate was already too high to detect a new plume due to ground-shine after a large amount of radionuclides was deposited on the grounds by precipitation.
4. Accordingly, it was firstly recognized that polluted air masses with high ^{137}Cs radioactivity (approximately 80 Bq m^{-3}) were present in Nakadori for more than half a day from the evening of March 20 to the morning of March 21, 2011.

5. An area with relatively high ^{137}Cs deposition by precipitation was observed on the morning of March 21, 2011 in the Tokyo metropolitan area, according to the airborne measurements by the MEXT[8] (Figure 1a). Within the area, time-integrated atmospheric ^{137}Cs concentrations at several SPM monitoring stations were also observed to be high due to the transport of a plume on March 21, 2011. However, in the Fukushima Prefecture, such a correlation was not clear.

In this study, large amounts of data, including more than twice the number of 40 SPM monitoring stations, have become available for the first time. Thus, we are preparing to comprehensively describe the atmospheric transport of radioactive materials in eastern Japan including discussions regarding the source term. In addition to our research, we expect many researchers to use the database presented in this study to produce new findings. In particular, we expect the uncertainty of internal dose rates from inhalation to decrease by the use of hourly release rates from radioactive materials from FDNPS, and of improved atmospheric transport models by validating the simulated results.

3.5. Scientific perspectives related to measuring radioactive nuclides in SPM samples. Because more than four years have passed, short-lived nuclides have decayed and can no longer be determined. However, in addition to radiocesium (^{134}Cs and ^{137}Cs), which were targeted in this study, several radioactive nuclides can be measured in the SPM samples. For example, ^{90}Sr and ^{129}I are representative, long-lived radionuclides. Unlike radiocesium, these radioactive nuclides cannot be determined by measuring gamma-rays. Thus, extra work and time are required for their measurement.

Because ^{90}Sr and ^{129}I are very important for assessing exposure dose due to internal exposure, the measurement of these nuclides is important. In particular, ^{129}I has been given considerable attention for estimating the internal exposure dose from short-lived radioiodine, ^{131}I. Because the half-life of ^{131}I is approximately 8 days, ^{131}I decays quickly. Therefore, the spreading behaviors of ^{131}I in the atmosphere from the nuclear reactors are not sufficiently understood, because the data of atmospheric ^{131}I concentrations by direct measurements in eastern Japan just after the accident were very limited. The concentrations of ^{131}I in the soil samples were measured in the Fukushima Prefecture as part of a project aimed at drawing a concentration map of radioactive nuclides that was led by MEXT in June-July, 2011.[16] However, because of the delayed initiation of this project, ^{131}I concentrations were observed only in areas where heavy deposition of radioactive nuclides, including ^{131}I, occurred.[17] Because ^{131}I and ^{129}I were fissiogenic nuclides from ^{235}U and released together into the environment by the FDNPS accident, ^{129}I can be used as a substitute for ^{131}I. Recently, the ^{129}I concentrations in soil samples collected in June-July 2011 were systematically determined using accelerator mass spectrometry (AMS). These results and a resulting concentration map were uploaded on the website.[18] Overall, these data can help to reveal how ^{129}I is presently distributed across a wider area than the area in which the ^{131}I deposition was directly measured by the MEXT project in June-July 2011.

However, several concerns exist regarding the use of ^{129}I data that were recently obtained from soil samples collected in 2011 to estimate the degree of internal exposure of ^{131}I. One major concern is whether it is possible to estimate the internal exposure dose from ^{131}I based on the presently determined ^{129}I data from soil samples. The ^{129}I activity in soil samples is an integrated value of the ^{129}I deposited on the ground from the atmosphere during the entire period, including the ^{129}I deposition before the Fukushima-nuclear-accident. If ^{129}I can be measured in the SPM samples used for this study, we will be able to estimate the internal exposure dose caused by ^{131}I based on such ^{129}I data more directly and reliably compared with estimations based on the ^{129}I deposition densities in the soil samples. To evaluate the internal exposure dose from ^{131}I, we must be able to answer the following questions with "yes": can ^{129}I be detected in the SPM samples and can ^{129}I be used in place of ^{131}I in the SPM samples? From our preliminary study, both questions were answered with yes. Next, we can set up a reliable scheme for estimating the internal exposure dose from ^{131}I based on ^{129}I measurements of the SPM samples used in this study.

4. Guides for using the ^{137}Cs radioactivity concentration data

There are two important issues to consider carefully when the data presented in this study are used as a primary dataset, namely, cross-contamination and detection limit. As described in Section 3.2, cross-contamination cannot be avoided in the SPM samples analyzed in this study. Careful attention should be given to the high concentration data in the major plumes from the SPM samples collected on PTFE filter tape. Regarding the detection limit, 3σ values are provided as a conventional method for presenting radioactivity measurement data. The detection limit depends on the experimental condition for gamma-ray counting. The guidelines below should be considered when using the radiocesium concentration data presented in this study.

(1) If there are more than one monitoring station located in a narrow area (2 to 3 km apart) and if both filter tapes of PTFE and GF are used in these monitoring stations, the data from the GF filter are more reliable, considering the higher probability of cross-contamination with the PTFE filter tape, as described in section 3.2. For example, four monitoring stations are located near in the Fukushima City (code #: 07-001, 07-002, 07-003 and 07-004). The PTFE filter tape was used only at the 07-002 station, and the GF filter tape was used at the other three stations. Therefore, the data from these three stations may be suitable as a primary data source. Consequently, in our previous paper[9] the data from these three stations with the use of GF were used. As another example, at the two monitoring stations in Tokyo (Himonya and Kakinokizaka, code#: 13-023 and 13-025, respectively), PTFE filter tape was used at station 13-023 and GF filter tape was used at station 13-025. Cross-contamination with these two stations was discussed in detail in Section 3.2, and the only data from station 13-025 were used in our previous paper.[9]

(2) At this stage, we introduce a simple method for determining the cross-contamination to users. For a given SPM station A where the PTFE filter tape was used, the ^{137}Cs concentration in a sampled spot that is located one round after the previously sampled spot with a high ^{137}Cs concentration around the peak time of a plume, might be considered unreasonably or irregularly high because these data were thought to be affected by cross-contamination. Then, the data were compared with those measured simultaneously at another SPM station B, that was located near SPM station A and where GF filter tape was used. In cases where the ^{137}Cs concentrations were nearly equal between the two SPM stations, the effects of cross-contamination can be neglected. However, when the ^{137}Cs concentration at A was higher than that at B and/or its temporal variation was not similar to that at B, the data were judged to be affected by cross-contamination and were not used in the previous paper.[9] In this case, because the ^{137}Cs concentration at A around the peak time was underestimated, the ^{137}Cs concentration derived for cross-contamination by calculating the contribution described in Section 3.2 is to be added for further study and quantitative discussion.

(3) The low ^{137}Cs concentrations (approximately 1 Bq m^{-3} or lower) measured at a SPM station using the PTFE filter tape after high concentrations at the peak time, may have been overestimated due to cross-contamination. This finding is evident when considering the data from the four sampling sites in the Fukushima City. Specifically, the ^{137}Cs concentrations at station 07-002, which used PTFE filter tape, were systematically higher than those at the other three stations that used GF filter tape at 9:00 and thereafter on March 21 (Figures 6d and 6e). Hence, comparisons between the simulated results by atmospheric transport models and measured data should be made carefully when the ^{137}Cs concentrations at the SPM stations with PTFE filter tapes are low (approximately 1 Bq m^{-3} or lower).

(4) Even when the concentration values are given by numerical values, it is acceptable to use such values as upper limits when the values are near the detection limit. This method is also acceptable when the concentration data are accompanied by large uncertainties (approximately 50% or higher for relative uncertainties).

The data reported in this paper are from only 99 of approximately 400 SPM sites, and radioactivity measurements are ongoing for the remaining samples. These data will be summarized and reported elsewhere and the ^{129}I measurement results will be reported in the near future.

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Appendices A and B are available from the journal homepage.