Bull Environ Contam Toxicol (2012) 89:368–375
DOI 10.1007/s00128-012-0659-y

Regional Air Pollution Caused by Dioxins from Numerous Emission Sources: Lessons from a Domestic Experience in Japan

Kazuyuki Suzuki · Katsuya Kawamoto

Received: 8 January 2012 / Accepted: 9 April 2012 / Published online: 1 May 2012

Abstract In this study, a large-scale field study was performed in order to distinguish between the contribution of the municipal solid waste incinerator and small clustered industrial waste incinerators in Fuchu city. The dioxin concentrations when only the municipal solid waste incinerator was being operated were found to range from 0.047 to 0.090 pg TEQ/m^3. The dioxin concentrations when only the clustered small industrial waste incinerators were being operated ranged from 0.085 to 0.25 pg TEQ/m^3. The concentrations in ambient air were more strongly affected by the clustered industrial waste incinerators than the municipal solid waste incinerator. Furthermore, the predicted concentrations by an atmospheric dispersion simulation model were consistent with the measured concentrations. From these results, the dioxin concentrations in ambient air were attributed primarily to the clustered small industrial waste incinerators.

Keywords Dioxins · Municipal solid waste incinerator · Clustered industrial waste incinerators · Atmospheric dispersion simulation model

Public concern about dioxin emissions from incinerators became particularly heightened in the 1990s in Japan. Dioxins are known to be highly toxic contaminants formed during combustion processes such as those of municipal solid waste incinerators (MSWIs). According to an inventory of dioxin emissions in Japan, the amounts of dioxins emitted from MSWIs, industrial waste incinerators, and small waste incinerators were 5,000, 1,500, and 700–1,153 g TEQ/year[1] in 1997, respectively, or about 65 %, 20 %, and 10 % of total emissions in Japan (Ministry of the Environment, Japan 2010). Therefore, waste incinerators were identified as a major source of dioxin pollution and there are many waste incinerators in Japan. However, the amount of dioxins emitted from incinerators has been decreasing gradually over the last decade (Ministry of the Environment, Japan 2010). The Law Concerning Special Measures against Dioxins in Japan enacted to reduce dioxin emissions, went into effect in 2000. Along with other standards and guidelines, the law established dioxin emission standards for waste incinerators and the Japanese ambient air quality standard for dioxin (0.6 pg TEQ/m^3).

Fuchu City, in Hiroshima prefecture, western Japan, encountered a dioxin problem in early 1999. The measured polychlorinated dibenzo-p-dioxins and dibenzofurans (PCDD/Fs) concentrations in the air in residential areas near the industrial area of the Motoyama district of the city exceeded the tentative guideline value (0.8 pg TEQ/m^3) applicable at that time. This significantly increased the anxiety of local citizens. An old-fashioned batch-type MSWI and many industrial waste incinerators were in use in the region. Because the town is well known for its woodcrafts, many woodworkers and furniture manufacturers live and work there. Therefore, woody wastes, such as woodchips generated during production of woodcrafts, were being burned in incinerators. In regions that have clustered dioxin emission sources, identification of major dioxin emission sources is extremely important in order to take effective countermeasures against the pollution. A

K. Suzuki (✉)
Material Cycles and Waste Management Group, Center for Environmental Science in Saitama, 914 Kamitanadare, Kazo, Saitama 347-0115, Japan
e-mail: suzuki.kazuyuki@pref.saitama.lg.jp

K. Kawamoto
Research Center for Material Cycles and Waste Management, National Institute for Environmental Studies, Tsukuba, Japan

number of studies have measured the dioxin level in the environment surrounding MSWIs. However, it is difficult to identify the primary dioxin emission sources from an ordinary survey, when numerous dioxins emission sources exist (like Fuchu city).

In this study, a large-scale field study was conducted to investigate which emission sources make significant contributions to regional pollution. Dioxin concentrations in ambient air were measured while controlling operation conditions of the two incinerator groups (the MSWI and clustered IWIs. Until now, there was no study that the large-scale field study like this was performed to identify source emission. Furthermore, the impact from the MSWI group and the clustered IWIs was evaluated using an atmospheric dispersion simulation model.

Materials and Methods

The study area (Fig. 1) was located in Fuchu, Hiroshima prefecture, western Japan. The city is well known for fine furniture manufacturing as a traditional indigenous industry. The district encompasses Motoyama Industrial Park, where many factories related to furniture manufacturing

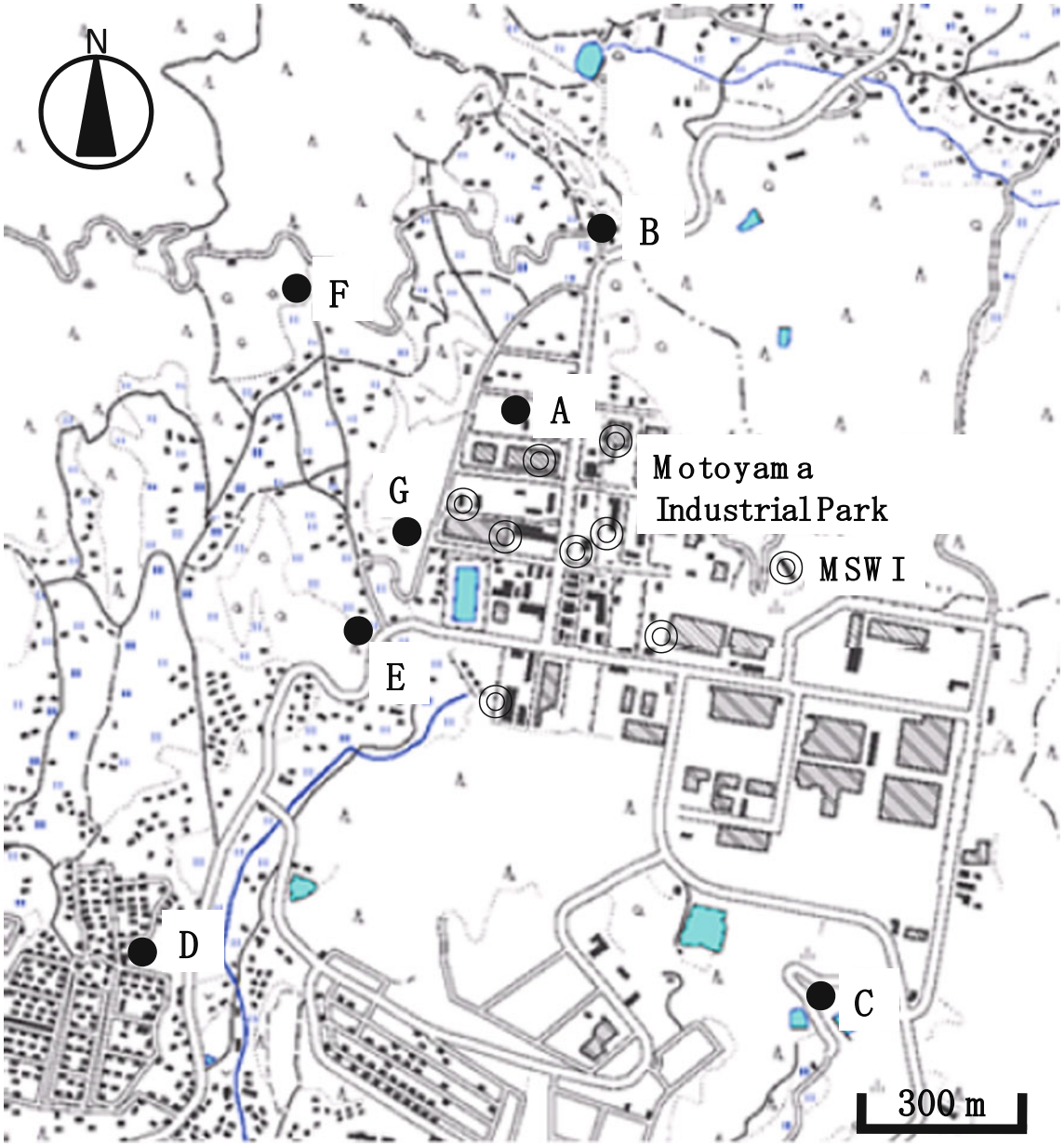

Fig. 1 Estimated dioxin emission sources and ambient air sampling sites in the study area (*double circle* emission source, *black circle* sampling site)

are located. In the factories, wastes generated from furniture manufacturing processes were burned in incinerators. There were also incineration plants operated by industrial waste disposal businesses. These incinerators, which acted as dioxin emission sources, were clustered in the industrial park. In addition, an MSWI with a capacity of 50 t/day was located in the vicinity, east of the industrial park. The MSWI was operated 8 h/day from 8 a.m. to 5 p.m. These incinerators were situated within an area of 1,200 m^2. The flue gases, including dioxins, were emitted from the incinerator stacks to the atmosphere every day. The locations of these emission sources are shown in Fig. 1. The subject area includes both an industrial area and a residential area. Ambient air was sampled at seven sites, A, B, C, D, E, F and G as shown in Fig. 1.

To elucidate the dioxins contribution of the MSWI, and the cluster of IWIs in the Motoyama Industrial Park to the concentration of dioxin in the ambient air of the study area, two groups of incinerators were operated to produce the following three conditions:

a. Both the clustered IWIs and the MSWI were completely shut down on January 1, 2000.
b. The operation of the MSWI was restarted on January 2, 2000. The clustered IWIs were not restarted until January 3, 2000.
c. The clustered IWIs were operated while the MSWI was shut down from April 25–26, 2000.

Ambient air measurement was conducted at sites A, B and C for all the field experiments noted above.

To assess the state of air pollution with regard to dioxins, long-term ambient air monitoring, including the monitoring done for these experiments, was conducted seasonally from January 1999 to January 2005.

Apart from the measurement, dioxin concentration data from the field experiments was assessed using an air dispersion model software package (Kakusansuketto ver. 1.0, provided by Hitachi Engineering & Services Co., Ltd., Tokyo, Japan). This model was designed based on the assumptions of steady-state emissions and dispersion of air pollutants with no chemical or photoreactions in the atmosphere. Meteorological parameters, including temperature, atmospheric stability, mixing height, wind speed, and wind direction, were prepared using data from the annual report of the Japan Meteorological Agency CD-ROM (Japan Meteorological Agency 2000a) and the Automated Meteorological Data Acquisition System (AMEDAS) annual report CD-ROM (Japan Meteorological Agency 2000b). Dioxin concentrations in the ambient air were calculated using data from the numerical map released by the Geographical Survey Institute, taking into account the effects of topography. The modeling results were used to evaluate the relative contributions of the two incinerator groups on ambient dioxin concentrations, but not for direct comparison between measured and modeled concentrations.

Ambient air sampling was performed using a single high-volume sampler 1.3 m high, starting in January 1999, according to Notification No. 68 of the Environment Agency (Environment Agency, Japan 1999a). The sampler consisted of a glass fiber filler (GFF) followed by a pre-washed polyurethane foam (PUF) plug connected to a suction pump. The PUF plug was spiked with 1 ng of $^{13}C_{12}$-labelled Surrogate Standard before field sampling. The flow rate was set at 0.7 m^3/min, allowing for sampling of about 1,000 m^3 of air each 24 h. Starting in March 2002, however, ambient air sampling was performed according to the Manual on Determination of Dioxins in Ambient Air (Ministry of the Environment, Japan 2001). Air sampling was done for 7 days with a high-volume sampler. Since much of the dioxin was contained in particulate matter, gaseous specimens were measured separately from particulates. Particulate substances in the ambient air were collected on the GFF. The gaseous components were collected on the PUF. The GFF and PUF were spiked with $^{13}C_{12}$-labeled internal standards and were then Soxhlet-extracted separately. The extract was spiked with $^{13}C_{12}$-labeled alternative standard and was then cleaned using acid–base extraction and multilayer silica gel and basic alumina columns. The final extract was concentrated and spiked with $^{13}C_{12}$-labeled recovery standard before analysis. The 2,3,7,8-chlorine-substituted congeners and the homologs of each chlorination class were detected using high-resolution gas chromatography/high-resolution mass spectrometry (HRGC/HRMS). The HRGC unit was equipped with a DB-5 MS fused silica capillary column (60 m, 0.25 mm i.d., 0.25 μm film thickness) and splitless injection. Helium was used as a carrier gas. The HRMS unit was equipped with a positive electron impact (EI+) source. The analyzer mode of the selected ion monitoring (SIM) was used at a resolution of 10,000. The toxicity equivalent (TEQ) values for PCDD/Fs and dioxin-like PCBs were calculated by using toxic equivalent factors (TEFs) re-evaluated by the World Health Organization in 1998 ($WHO\text{-}TEQ_{1998}$).

Results and Discussion

Figure 2 shows the ambient air dioxin measurement data from the experiments when the operation of the MSWI of Fuchu and clustered IWIs in Motoyama Industrial Park were controlled. The atmospheric concentrations of dioxins at sites A, B and C in the measurement made when both the MSWI and the clustered IWIs were stopped were 0.014, 0.016 and 0.022 pg TEQ/m^3, respectively. These concentrations were regarded as background levels at these sites.

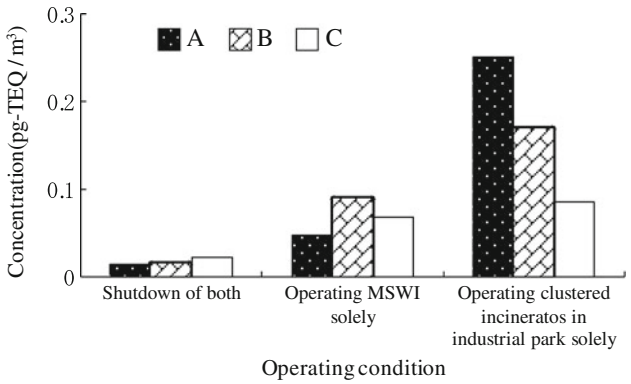

Fig. 2 Dioxin concentrations in ambient air at three sites under three experimental incinerator operational conditions

The concentrations at sites A, B and C when only the MSWI was in operation were 0.047, 0.090 and 0.068 pg TEQ/m^3. These levels were 3.4, 5.6 and 3.1 times higher, respectively, than when both the MSWI and the clustered IWIs were stopped. In contrast, atmospheric dioxin concentrations measured when only the clustered incinerators were operated were 0.25, 0.17 and 0.085 pg TEQ/m^3, respectively, which were 18, 11 and 3.9 times higher than the background levels. These results suggest that the cluster of incinerators in the Motoyama Industrial Park was the dominant dioxin emission source. In particular, the increase in dioxin concentrations at site A was especially high between the second (MSWI only) and third (clustered IWIs only) experiments. Hence, emitted gases from the IWIs in the Motoyama Industrial Park strongly affected the atmospheric dioxin concentration at site A because this site was located near the northern edge of the Motoyama Industrial Park. The dioxin concentrations in the particulate-phase at sites A, B and C were 59 %, 51 % and 48 %, respectively. These were higher than the other two experimental conditions (17 %–33 %) except for site C when all emission sources were shut down. When only the MSWI was operated, dioxins in the particulate phase of the flue gas from the MSWI were about 40 % lower. These results indicate that the flue gas from the clustered IWIs in the Motoyama Industrial Park had high levels of particulate dioxins, and their nature might affect the regional polychlorinated dibenzo-p-dioxins and dibenzofurans (PCDD/F) air concentration profile. The PCDD/F homolog distribution between gas and particulate components is also of interest. T_4CDDs and T_4CDFs were the most abundant homologs in the gas phase, and the relative ratio of each homolog to the total concentration decreased with increased chlorine substitution number for both PCDDs and PCDFs. The high gaseous concentrations of T_4CDDs and T_4CDFs suggested that the less-chlorinated compounds distributed abundantly to the gas phase because

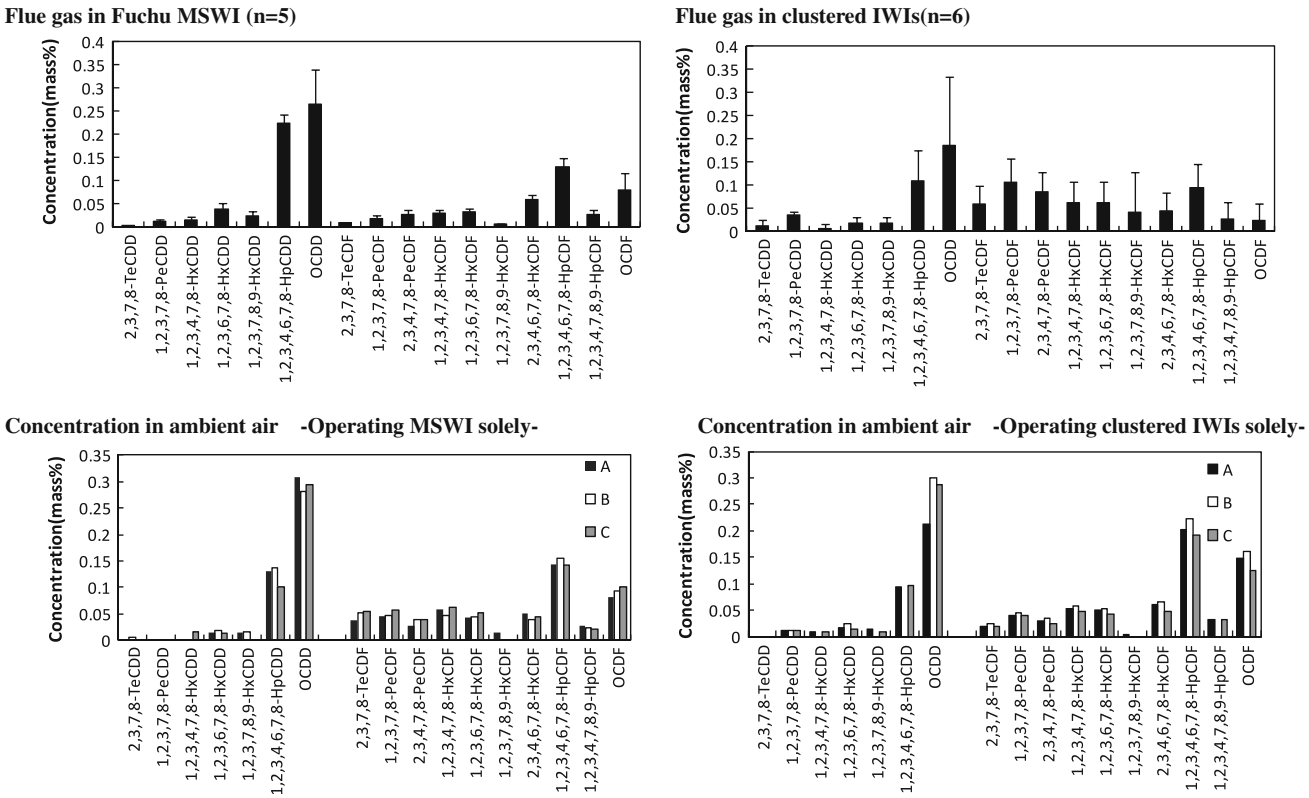

Fig. 3 Congener profiles of seventeen 2,3,7,8 substituted PCDD/Fs obtained from each samples (*error bar* represents the standard deviation)

Table 1 Operational parameters and characteristics of emission sources clustered in the city of Fuchu

	Emission gas				Stack		Remarks
	Dioxins (ng TEQ/m^3_N)	Gas volume (m^3_N/s^1)	Discharge rate (m/s^1)	Temperature (°C)	Height (m)	Radius (m)	
MSWI in Fuchu City	8.8	10	8.1	254	59	1.2	MSWI was abolished in November 2002
Motoyama industrial park							
Establishment 1	29	4.361	28	147	10.19	0.55	
Establishment 2	0.025	0.528	7.5	180	10	0.39	Incinerator was abolished in April 2001
Establishment 3	71	1.506	21.1	131	10	0.4	Incinerator was abolished in December 2002
Establishment 4	0.84	5.028	17	57	5.95	0.68	Incinerator was abolished in December 2002
Establishment 5	0.22	0.333	5.2	173	12	0.36	Now defunct.
Establishment 6	37	0.139	4.7	333	5.98	0.3	Incinerator was abolished in January 2001
Establishment 7	2.9	1.9	13.5	245	25	(0.5)	Factory closed in 2001
Establishment 8	0.32	4	16.9	72	12	(0.5)	
Establishment 9	0.095	0.45	17.6	239	4.69	0.25	Incinerator was abolished in December 2002

Parentheses show hypothetical stack radii where data could not be obtained

of their relatively high vapor pressures. However, no simple rule was found for the concentrations in the particle phase, probably because there were rather high concentrations of moderately chlorinated homologs with five and six chlorines that were predominantly distributed in particulates. Fig. 3 shows the congener profiles of seventeen 2,3,7,8 substituted PCDD/Fs obtained from each samples. The congener profiles of flue gas in MSWI was characterized by comparatively high abundance of OCDD, 1,2,3,4,6,7,8-HpCDD, 1,2,3,4,6,7,8-HpCDF and OCDD. The congener profile was some equal to that of flue gas in the clustered IWIs. However, the chemical composition in IWIs varied considerably as shown in Fig. 3. The congener profile of both emission sources was similar to that of both concentrations in ambient air measured when only MSWI was operated and only the clustered IWIs were operated. From the result, it is impossible for the analysis of seventeen 2,3,7,8 substituted congener profile to identify the dioxins emission sources.

A Gaussian plume/puff dispersion mode was used to determine the long-term (seasonal and year value) quasi-stable dioxin concentrations that could be expected under the same conditions as the field experiment. The model was calculated using the measured emission data shown in Table 1 that covers the MSWI and the clustered IWIs in the Motoyama Industrial Park. Fig. 4 shows the predicted atmospheric dioxin concentrations in the area investigated on a seasonal average basis. If only the MSWI was operated, the maximum dioxin concentration appeared at two points: about 2 km north–northwest and about 1 km east of the MSWI. The predicted maximum dioxin concentration in ambient air was 0.05 pg TEQ/m^3. The calculations indicated no seasonal variation for conditions similar to the field experiments. Because the MSWI's stack was 59 m high, dispersion of pollutants produced lower atmospheric dioxin concentrations in this area. These results demonstrated that the MSWI was not a significant source of dioxins in this study area. When the clustered IWIs in Motoyama Industrial Park were operated alone, the modeled maximum dioxin concentration appeared in the vicinity east of the clustered incinerators. The predicted maximum dioxin concentration was 0.91 pg TEQ/m^3. The predicted dioxin concentrations at site A ranged from 0.10 to 0.30 pg/m^3. The calculated results agreed well with the

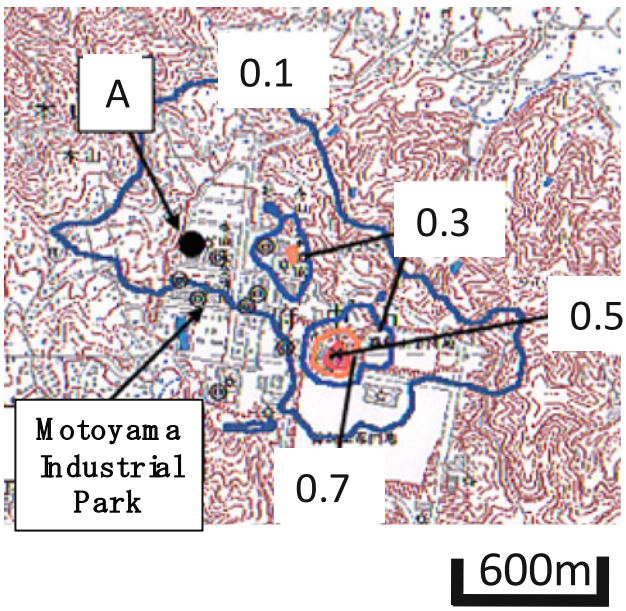

Fig. 4 Predicted dioxin concentrations in the targeted area using a dispersion model (unit pg TEQ/m^3)

Table 2 Dioxin concentration changes in ambient air at seven sites

	1999				2000			2001				2002			
	Winter	Spring	Summer	Autum	Winter	Spring	Autum	Winter	Spring	Summer	Autum	Winter	Spring	Summer	Autum
A	2.0	1.7	0.35	0.42	0.082	0.59	0.90	0.12	0.19	0.059	0.11	0.066	0.047	0.058	0.075
B			0.18	0.32	0.062	0.38	0.11	0.096	0.21	0.041	0.12	0.16			
C			0.072	0.087	0.018	0.26									
D			0.13	0.076	0.25	0.13									
E							0.62	0.11	0.093	0.037	0.13	0.084			
F							0.082	0.029	0.063						
G										0.044	0.13	0.13	0.046	0.071	0.09

	2003				2004				2005
	Winter	Spring	Summer	Autum	Winter	Spring	Summer	Autum	Winter
A	0.14	0.068	0.054	0.094	0.25	0.32	0.24	0.22	0.25
B									
C									
D									
E									
F									
G	0.23								

Unit: pg TEQ/m^3_N

measured dioxin concentration of 0.25 pg TEQ/m^3. These results support the hypothesis that the clustered IWIs in the Motoyama Industrial Park were the main dioxin source, not the MSWI.

Table 2 shows dioxin concentration trends in ambient air at seven monitoring sites from January 1999 to March 2005. As the figure shows, the atmospheric dioxin concentrations at site A, in the vicinity of the Motoyama Industrial Park, were more than twice the tentative guideline value of 0.8 pg TEQ/m^3 that was applicable from January to April 1999. This situation was the problem that this study was undertaken to investigate. The dioxin concentrations at site A decreased after the investigation began, but rose in the spring and autumn in 2001. In particular, monitoring data in the autumn showed dioxin concentration exceeding the Japanese ambient air quality standard (JAQS) of 0.6 pg TEQ/m^3. After that, the concentration again decreased, achieving a level considerably lower than the JAQS. The average dioxin concentrations at site A in 2002 and 2003 were very low, 0.0072 and 0.082 pg TEQ/m^3, respectively. The monitored concentrations were comparable to those in a rural and remote areas in United States (−0.01 and −0.001 pg TEQ/m^3) (Cleverly et al. 2007), the mean concentration in background/control site of Catalonia (0.012 pg WHO-TEQ/m^3) (Mari et al. 2008), those measured in a rural area in Italy (0.02–0.05 pg TEQ/m^3) (Caserini et al. 2004) and to background levels in Japan (0.013 pg TEQ/m^3) (Environment Agency, Japan 1999b). It is thought that strict regulation of incineration flue gas drastically reduced dioxin emissions and lowered their concentrations in ambient air. It is interesting to note, from Table 2, that site A's dioxin concentrations were the highest of all sites. Because site A was very close to the Motoyama Industrial Park, which was considered to be the main source of dioxin emissions, dioxin concentrations were often rather high, because of the area's meteorological conditions. In particular, site A was downwind of the clustered IWIs in Motoyama industrial park because the most common winds at site A were south–southwestward (14.1 %). These results suggest that dioxin emission sources that are located at south of site A might contribute to its high dioxin concentration. Ambient air dioxin concentrations at site A in 1999 were higher than levels in the vicinity of incinerators in Korea (0.032–0.965 pg TEQ/m^3) (kim et al. 2008), Southern Taiwan (0.028–0.136 pg TEQ/m^3) (Wu et al. 2009), northeast Italy (average 0.02 ± 0.01–0.16 ± 0.02 pg TEQ/m^3) (Colombo et al. 2009), Japan (0.021–0.26 pg TEQ/m^3) (Takei et al. 2000) and Portugal (0.125–0.247 pg TEQ/m^3) (Coutinho et al. 2000) and comparable to that in China (0.059–3.03 pg TEQ/m^3) (Xu et al. 2009).

The Law Concerning Special Measures against Dioxins that has been in effect since December 2002 tightened regulations regarding dioxin exhaust emissions from incinerators. As a result, several incinerators in the Motoyama Industrial Park were abolished, as shown in Table 1. Currently, only one incinerator, which is regulated by the law, is operated in the industrial park. The dioxin concentrations of emitted gas from the incinerator were a lower level (0.53–2.4 ng TEQ/m^3) between 2002 and 2010 (16). Moreover, the municipal solid waste incinerator in Fuchu City was abolished. In its place, a plant to convert municipal solid waste to refuse-derived fuel (RDF) was constructed, which began operations in November 2002. The RDF was carried to an RDF incinerator that generates electricity in Fukuyama City, 20 km away from Fuchu City. Consequently, atmospheric dioxin concentrations in Fuchu City have continued to be low. In this regional experience, locating an industrial area and a residential area near each other increased the anxiety of the residents of the district. To reduce this anxiety, it is important to confirm the quantitative influence of present and future emission sources. Comprehensive analyses of this study present a useful approach for identifying the major dioxin pollution sources that are responsible for the deterioration of the air quality of local areas. The experience in this region can be applied to issues of atmospheric semi-volatile compounds, such as dioxin pollution from clustered emission sources in developing and developed countries.

Acknowledgments The authors thank the Fuchu City for its kind support and cooperation.

References

Caserini S, Cernuschi S, Giugliano M, Grosso M, Lonati G, Mattaini P (2004) Air and soil dioxin levels at three sites in Italy in proximity to MSW incineration plants. Chemosphere 54: 1279–1287

Cleverly D, Ferrario J, Byrne C, Riggs K, Joseph D, Hartford P (2007) A general indication of the contemporary background levels of PCDDs, PCDFs, and coplanar PCBs in the ambient air over rural and remote areas of the United State. Environ Sci Technol 41:1537–1544

Colombo A, Benfenati E, Mariani G, Lodi M, Marras R, Rotella G, Senese V, Fattore E, Fanelli R (2009) PCDD/Fs in ambient air in north-east Italy: the role of a MSWI inside an industrial area. Chemosphere 77:1224–1229

Coutinho M, Ferreira J, Gomes P, Borrego C (2000) Atmospheric levels of PCDD/PCDF during the test phase of a municipal solid waste incinerator, in Portugal. Organohalogen Compd 46: 447–450

Environment Agency, Japan (1999a) Notification No. 68 of Environment Agency

Environment Agency, Japan (1999b) Results of an urgent simultaneous nationwide survey of dioxins (conducted in 1998). http://www.env.go.jp/en/chemi/dioxins/urgent_Nation-wide_survey.pdf#search='urgentdioxinssurvey'

Japan Meteorological Agency (2000a) Annual report of the Japan Meteorological Agency CD-ROM

Japan Meteorological Agency (2000b) Automated meteorological data acquisition system (AMEDAS) annual report CD-ROM

Kim KS, Shin SK, Kim KS, Song BJ, Kim JG (2008) National monitoring of PCDD/DFs in environmental media around incinerators in Korea. Environ Int 34:202–209

Mari M, Nadal M, Schuhmacher M, Domingo JL (2008) Monitoring PCDD/Fs, PCBs and metals in the ambient air of an industrial area of Catalonia, Spain. Chemosphere 73:990–998

Ministry of the Environment, Japan (2001) Manual on determination of dioxins in ambient air. http://www.env.go.jp/en/chemi/dioxins/manual.pdf#search='method%20manual%20for%20determination%20of%20dioxins%20in%20ambient%20air'

Ministry of the Environment, Japan (2010) Dioxin Emission Inventory. http://www.env.go.jp/air/report/h22-09/index.html

Takei T, Araki S, Kanai M, Morita M (2000) Enviromental levels of dioxins: results of nationwide survey of dioxins. Organohalogen Compd 46:475–478

Wu YL, Lin LF, Hsieh LT, Wang LC, Chang-Chien GP (2009) Atmospheric dry deposition of polychlorinated dibenzo-p-dioxins and dibenzofurans in the vicinity of municipal solid waste incinerators. J Hazard Mater 162:521–529

Xu MX, Yan JH, Lu SY, Li XD, Chen T, Ni MJ, Dai HF, Wang F, Cen KF (2009) Concentrations, profiles, and sources of atmospheric PCDD/Fs near a municipal solid waste incinerator in eastern China. Environ Sci Technol 43:1023–1029