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Chemosphere 78 (2010) 127–133

Contents lists available at ScienceDirect

Chemosphere

journal homepage: www.elsevier.com/locate/chemosphere

Characteristics of the abundance of polychlorinated dibenzo-*p*-dioxin and dibenzofurans, and dioxin-like polychlorinated biphenyls in sediment samples from selected Asian regions in Can Gio, Southern Vietnam and Osaka, Japan

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ARTICLE INFO

Article history:
Received 12 June 2009
Received in revised form 1 October 2009
Accepted 1 October 2009
Available online 4 November 2009

Keywords:
Agent Orange
Can Gio
Commercial PCBs
Incineration of solid wastes
Natural origin
Osaka

ABSTRACT

The levels of polychlorinated dibenzo-*p*-dioxin and dibenzofuran (PCDD/PCDFs), and dioxin-like polychlorinated biphenyls (DL-PCBs) were determined in sediment samples from Can Gio, South Vietnam, and Osaka, Japan. Can Gio is known for the defoliation of its mangrove forests by aerial spraying with Agent Orange during the Vietnam War, whereas Osaka is renowned for a PCDD/PCDF pollution accident at a municipal solid-waste incinerator. For comparison, we also analyzed PCDD/PCDFs and DL-PCBs in sediment samples from Hue and Hanoi, Vietnam. The toxic equivalent quantity (TEQ) values in Can Gio were as high as those in Hue, Hanoi, and suburban areas of Osaka, but much lower than those in urban areas of Osaka. The proportion of the World Health Organization (WHO)-TEQ value contributed by 2,3,7,8-tetrachlorinated dibenzo-*p*-dioxin (TCDD) in Can Gio was approximately 30%, higher than the values in the other sample areas. These data suggest that residual sedimentary TCDD that originated from aerial spraying of Agent Orange occur in only low concentrations in Can Gio. The main contributors to WHO-TEQ values in Can Gio are natural sources, as in Hue. In contrast, commercial PCBs are the dominant contributors to WHO-TEQ values in Hanoi. In Osaka, agrochemicals used in rice cultivation, the incineration of solid waste, and commercial PCBs equally contributed to WHO-TEQ values at suburban locations. The dumping of incinerator-related materials and/or the inadequate management of commercial PCBs have resulted in significantly elevated WHO-TEQ values of 240–370 ng kg^{-1} dw at urban locations in Osaka.

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1. Introduction

It is recognized worldwide that the herbicide mixtures sprayed aerially during the Vietnam War are harmful to human health, resulting in, for example, birth defects. The well-known herbicide "Agent Orange" is a mixture of 2,4-dichlorophenoxyacetic acid (2,4-D) and 2,4,5-trichlorophenoxyacetic acid (2,4,5-T). A congener, 2,3,7,8-tetrachlorinated dibenzo-*p*-dioxin (TCDD), was a contaminant in the 2,4,5-T portion of Agent Orange. Therefore, previous research has focused on its contamination of human and organic tissues (Schecter et al., 2001; Dwernychuk et al., 2002), as well as of soil and sediments from limited areas in Vietnam (Dwernychuk et al., 2002; Mai et al., 2007).

Can Gio, located upon the Mekong delta approximately 100 km south of Ho Chi Minh City (HCMC), is also a 'hot spot' of herbicide use and is known internationally for the reestablishment of its mangrove forests (Hong, 1996). As a result of aerial spraying with herbicides by US forces, 40 000 ha of mangrove forests was defoliated, leaving just 4500 ha remaining. Since the conflict, several projects have been conducted to restore the mangroves, and 20 000 ha of mangrove forests have now been reestablished. However, the presence of residual TCDD in this region has not been investigated.

Regarding other Asian regions, large amounts of polychlorinated dibenzo-*p*-dioxin (PCDD), polychlorinated dibenzofuran (PCDF), and dioxin-like polychlorinated biphenyls (DL-PCBs) had been emitted into the environment in Japan until the early 2000s

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0045-6535/$ - see front matter © 2009 Elsevier Ltd. All rights reserved.
doi:10.1016/j.chemosphere.2009.10.003

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(Ministry of the Environment [MOE], Government of Japan, 2006a), more than that emitted in the main European countries (The United Nations Environment Programme [UNEP], 1999). The main PCDD/PCDF sources in Japan are reported to be herbicides used for rice production, such as pentachlorophenol (PCP) and chloronitrofen (CNP), and the incineration of solid wastes (Yasuhara et al., 1987; Masunaga et al., 2001a,b). In the latter case, most solid wastes in Japan are disposed of at municipal solid-waste incinerators (MSWIs). In the 1990s, an abnormally high toxic equivalent quantity (TEQ) value of 8500 ng kg^{-1} dw was detected in soils around an MSWI in Nose, north of Osaka (MOE, Government of Japan, 2006b). Since this incident, the levels of dioxins in river water, sediments, etc. in Japan have been monitored by local government, and the TEQ values have often exceeded the Japanese environmental criteria for PCDD/PCDFs and DL-PCBs in sediments and river water (150 ng kg^{-1} dw and 1.0 pg L^{-1}, respectively), especially in urban and industrial areas of cities such as Osaka and Tokyo (MOE, Government of Japan, 2006b).

The basin of the Kanzaki River is typical of areas in Osaka affected by PCDD/PCDF and DL-PCB pollution in sediments and river water. The Kanzaki River runs through the north of central Osaka, which is a mixed industrial, trade, and residential area. There are many incineration facilities in this basin. Therefore, TEQ values exceeding the environmental criteria are likely to be strongly associated with incineration-related sources, as in Nose. However, many monitoring data obtained by the Department of Environment, Agriculture, and Fisheries (DOEAF), Osaka Prefectural Government (2009), have not been effectively utilized to advance our understanding of the distribution and source of PCDD/PCDFs and DL-PCBs.

In the present study, to understand the characteristics of PCDD/PCDF and DL-PCB pollution in the surface sediments of different parts of Asia, we selected two regions for analysis: the mangrove forests in Can Gio, South Vietnam, and the basin of the Kanzaki River in Osaka, Japan. For comparison, sediment samples were also collected from rural sites in Hue, central Vietnam, and from urban sites in Hanoi, the capital and second-largest city of Vietnam.

2. Experimental

2.1. Sampling

The 10 sampling locations in Can Gio are shown in Fig. 1. Details of the sampling locations in Hue, Hanoi, and Osaka are given in Table 1. The sediment samples from Can Gio were collected at more than 10 points near the sampling site, using a steam shovel on 12 January 2004, when seven samples (Nos. 1–7) were collected, and on 26 October 2004, when three samples (Nos. 8–10) were collected. Samples from other regions were collected at more than three points near the sampling site, using an Eckmann dredge sampling apparatus. The sediment samples from Vietnam and Osaka were placed in chemically cleaned aluminum bags and glass vessels, respectively, and then transported to a laboratory in Osaka, Japan. The transported sediment samples were dried under moderate conditions in the laboratory.

2.2. Sample analysis

The levels of PCDD/PCDFs and DL-PCBs in the sediment samples were determined using the method provided by MOE, Government

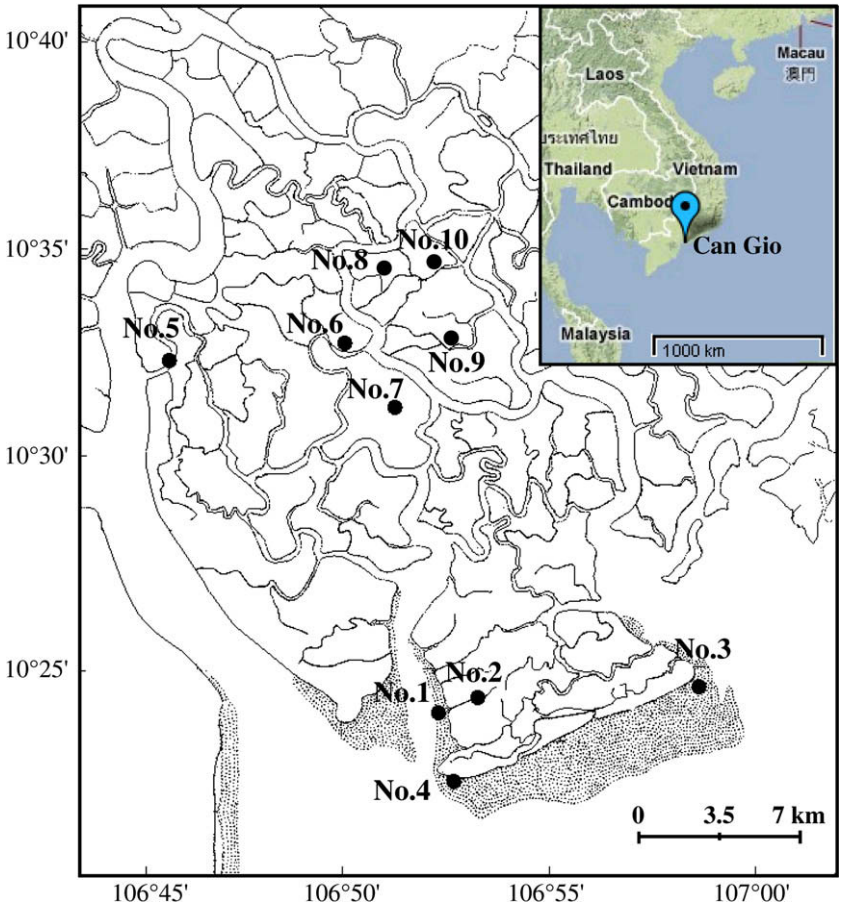

Fig. 1. Map showing sampling locations in the mangrove forests of Can Gio, South Vietnam.

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Table 1
Details of sampling locations in Hue and Hanoi, Vietnam and suburban and urban locations in Osaka, Japan.

Site No.	City	Location	Description	Geographical coordination		Sampling date
				North	East	
11	Hue	LangCo Lagoon	Rural	16°33′37″	107°37′15″	13/Jan./2003
12	Hue	ThuyTu Lagoon	Rural	16°29′30″	107°43′20″	12/Jan./2003
13	Hue	CauHai Lagoon	Rural	16°20′19″	107°52′28″	10/Jan./2003
14	Hanoi	TrucBach Lake	Urban	21°02′46″	101°50′21″	27/Jul./2002
15	Hanoi	West Lake	Urban	21°03′27″	105°50′15″	27/Jul./2002
16	Osaka	R. Ibaraki	Suburb	34°49′45″	135°34′17″	07/Aug./2003
17	Osaka	R. Senri	Suburb	34°45′58″	135°27′06″	07/Aug./2003
18	Osaka	R. Taisho	Suburb	34°46′24″	135°33′27″	07/Aug./2003
19	Osaka	R. Yono	Suburb	34°50′27″	135°25′26″	07/Aug./2003
20	Osaka	R. Mino	Suburb	34°48′00″	135°25′51″	07/Aug./2003
21	Osaka	R. Katsuoji	Suburb	34°50′26″	135°33′15″	07/Aug./2003
22	Osaka	R. Samonden	Urban	34°42′42″	135°25′47″	14/Nov./2003
23	Osaka	R. Kanzaki	Urban	34°44′01″	135°27′32″	14/Nov./2003
24	Osaka	R.Ai	Urban	34°47′14″	135°34′55″	09/Sep./2003
25	Osaka	R.Ai	Urban	34°45′30″	135°31′55″	09/Sep./2003
26	Osaka	Banda Waterway	Urban	34°45′26″	135°32′24″	09/Sep./2003
27	Osaka	Banda Waterway	Urban	34°46′24″	135°33′47″	09/Sep./2003
28	Osaka	Banda Waterway	Urban	34°47′12″	135°34′56″	09/Sep./2003
29	Osaka	Ajifu Waterway	Urban	34°45′26″	135°33′08″	09/Sep./2003

of Japan (2000). Sediment sample was extracted with toluene for 16 h using a Soxhlet apparatus. After extraction, the 18 $^{13}C_{12}$-labeled PCDD/PCDF and 12 DL-PCB internal standards were added to the extract to check the recovery of the dioxin congeners throughout the clean-up procedure. The extract was concentrated to approximately 1 mL with solvent exchange to hexane, shocked with concentrated sulfuric acid, purified with silica by gel column chromatography and the addition of reduced copper, and then activated charcoal/silica by gel column chromatography. After the addition of three injection internal standards ($^{13}C_{12}$–1,3,6,8-TeCDF, $^{13}C_{12}$–1,2,3,4,6,8,9-HpCDF, and $^{13}C_{12}$–2,3′,4′,5-TeCB), each fraction was concentrated to 50 μL under a gentle stream of pure nitrogen gas. All $^{13}C_{12}$-substituted and native PCDD/PCDFs and DL-PCBs were purchased from Wellington Laboratories Inc. (Ontario, Canada). Solvents and reagents were dioxin-free analytical grade except for sulfuric acid (heavy metal analytical grade), as obtained from Wako Pure Chemical Industries Ltd. (Osaka, Japan) and Kanto Chemical Co., Inc. (Tokyo, Japan).

All samples were analyzed by high-resolution gas chromatography (GC)/high-resolution mass spectrometry (MS) (HP5890, Agilent, DE, USA; JMS-700D, JEOL, Tokyo, Japan; Kakimoto et al., 2006). The method detection limits (MDLs) were 0.06–0.28 ng kg^{-1} dw for Te–HpCDD/Fs and DL-PCBs, and 0.40 ng kg^{-1} dw for OCDD/Fs. The average recoveries were 90.5 ± 12.9% for 18 $^{13}C_{12}$-labeled PCDD/PCDF congeners and 100.0 ± 13.8% for 12 $^{13}C_{12}$-labeled DL-PCB congeners.

In this study, the WHO-TEQ value for each sample was obtained from the concentrations of 2,3,7,8-subsutituted PCDD/PCDFs and DL-PCBs using their toxic equivalency factors (TEFs) proposed by WHO in 1998.

3. Results and discussion

3.1. PCDD/PCDF concentrations in sediment samples from Vietnam and Osaka

The concentrations of the total PCDD/PCDF congeners (∑PCDD/PCDFs) are listed in Table 2. The average ∑PCDD/PCDF concentration in the sediment samples in Can Gio was as high as those in Hue, Hanoi, and suburban Osaka, but much lower than those in urban locations at Osaka. At urban locations in Osaka, extremely high ∑PCDD/PCDF concentrations were detected at locations 22, 23, 28, and 29 (13 000, 26 000, 19 000, and 12 000 ng kg^{-1} dw, respectively).

PCDD/PCDF concentrations in Can Gio, Hue, Hanoi, and suburban Osaka were as high as those reported previously in the Aluoi Valley (average: 550 ± 550 ng kg^{-1} dw; Dwernychuk et al., 2002) and suburban areas of Tokyo (average: 64 ± 56 ng kg^{-1} dw, Bureau of Environment [BOE], Tokyo Metropolitan Government, 2009), whereas those at urban locations in Osaka were the same as those in urban Tokyo (average: 4300 ± 7400 ng kg^{-1} dw, BOE, Tokyo Metropolitan Government, 2009).

3.2. Profiles of PCDD/PCDF homologues in sediment samples from Vietnam and Japan, and estimation of their sources

As listed in Table 2, the dominant homologues in Can Gio and Hue were OCDD, HpCDDs, and HxCDDs, in the order of OCDD > HpCDDs ≈ HxCDDs. The patterns in Hanoi were not completely consistent with those in Can Gio and Hue. The concentrations of PCDF congeners in Hanoi were higher than those in Can Gio and Hue. These data suggest that the main sources of PCDD/Fs in Can Gio are similar to those in Hue, but different from those in Hanoi.

The strong predominance of OCDD in the PCDD/PCDF profiles is considered to have mainly originated from pentachlorophenol (PCP; Baker and Hites, 2000; Masunaga et al., 2001a,b) or natural sources (Gadomski et al., 2004; Gaus et al., 2001). In general, PCP is used to enhance the growth of rice and forests. However, the Pesticide Action Network North America Update Service (PANUPS; 1995) reported that PCP has not been widely used for rice cultivation in Vietnam. Baker and Hites (2000) proposed that the strong predominance of OCDD and HpCDDs (with OCDD > HpCDDs) and the low content of other PCDD/PCDF homologues is attributable to the de novo synthesis (in the atmosphere) of PCP used for treating timber. However, the PCDD/PCDF patterns in Can Gio, Hue, and Hanoi are not in agreement with those attributable to the synthesis of PCP. Above all, the high occurrence of OCDD in Vietnam is not attributable to PCP.

In terms of naturally occurring OCDD, Gaus et al. (2001) reported that PCDD/PCDF profiles are characterized by low or undetectable PCDFs and specific 2,3,7,8-substituted HxCDD distributions. Table 2 shows that the PCDF concentrations in Can Gio and Hue were low or undetectable, except at location 7. In term of the 2,3,7,8-substituted HxCDD distributions, Gaus et al. (2001) reported that the ratios of 1,2,3,7,8,9-HxCDDs and 1,2,3,6,7,8-HxCDDs to the total 2,3,7,8-substituted HxCDDs from natural

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Table 2
Average concentrations of PCDD/PCDF and DL-PCB congeners and WHO-TEQ values in sediment samples from Can Gio, Hue, and Hanoi, Vietnam and Osaka, Japan

Congeners	Vietnam			Osaka	
	Can Gio (n = 10)	Hue (n = 3)	Hanoi[a] (n = 2)	Suburban locations (n = 6)	Urban locations (n = 8)
Concentration (ng kg^{-1} dw)					
PCDD congeners					
1,3,6,8-TeCDD	1.3 ± 1.6	0.86 ± 0.48	2.9 ± 2.8	21 ± 25	400 ± 300
1,3,7,9-TeCDD	1.1 ± 1.5	0.75 ± 0.48	1.6 ± 1.7	7.0 ± 8.3	190 ± 150
2,3,7,8-TeCDD	0.80 ± 0.42	0.29 ± 0.33	0.21 ± 0.05	0.08 ± 0.19	7.4 ± 9.5
TeCDDs	11 ± 7.6	15 ± 14	9.7 ± 0.42	34 ± 42	720 ± 510
1,2,3,7,8-PeCDD	0.60 ± 0.44	0.86 ± 0.75	0.52 ± 0.01	0.40 ± 0.77	20 ± 15
PeCDDs	17 ± 11	32 ± 33	11 ± 0.00	9.3 ± 14	500 ± 400
1,2,3,4,7,8-HxCDD	0.80 ± 0.27	1.3 ± 1.2	0.65 ± 0.08	0.41 ± 0.81	32 ± 27
1,2,3,6,7,8-HxCDD	1.3 ± 0.8	2.2 ± 2.1	1.3 ± 0.07	0.83 ± 1.5	62 ± 48
1,2,3,7,8,9-HxCDD	2.3 ± 1.6	4.2 ± 4.0	1.3 ± 0.07	0.69 ± 0.92	49 ± 39
HxCDDs	60 ± 32	140 ± 150	22 ± 0.71	11 ± 16	880 ± 840
1,2,3,4,6,7,8-HpCDD	18 ± 7.9	40 ± 41	18 ± 0.71	10 ± 13	580 ± 440
HpCDDs	62 ± 29	160 ± 180	47 ± 1.4	20 ± 26	1200 ± 910
OCDD	200 ± 88	650 ± 740	310 ± 7.1	120 ± 130	2900 ± 2200
total PCDDs	350 ± 160	980 ± 1100	390 ± 14	196 ± 231	6100 ± 4400
PCDF congeners					
2,3,7,8-TeCDF	1.5 ± 2.3	0.51 ± 0.29	6.1 ± 0.07	0.63 ± 1.3	24 ± 17
TeCDFs	17 ± 29	8.0 ± 4.1	58 ± 1.4	16 ± 34	540 ± 370
1,2,3,7,8-PeCDF	0.81 ± 1.2	0.51 ± 0.21	2.7 ± 0.07	1.2 ± 2.6	60 ± 49
2,3,4,7,8-PeCDF	1.10 ± 1.30	0.16 ± 0.28	2.2 ± 0.14	1.1 ± 2.7	70 ± 67
PeCDFs	6.8 ± 10	6.1 ± 2.5	31 ± 0.00	16 ± 33	840 ± 760
1,2,3,4,7,8-HxCDF	0.48 ± 0.29	0.33 ± 0.29	2.0 ± 0.00	1.5 ± 3.1	130 ± 130
1,2,3,6,7,8-HxCDF	0.49 ± 0.30	0.21 ± 0.36	2.0 ± 0.00	1.4 ± 3.2	120 ± 140
1,2,3,7,8,9-HxCDF	0.60 ± 0.55	0.09 ± 0.08	0.73 ± 0.06	0.08 ± 0.12	15 ± 16
2,3,4,6,7,8-HxCDF	N.D.[c]	0.18 ± 0.31	1.4 ± 0.35	1.4 ± 3.9	260 ± 330
HxCDFs	3.1 ± 2.1	4.8 ± 1.6	16 ± 0.71	17 ± 38	1400 ± 1500
1,2,3,4,6,7,8-HpCDF	2.3 ± 1.3	2.6 ± 1.0	7.2 ± 0.28	7.1 ± 15	850 ± 1000
1,2,3,4,7,8,9-HpCDF	2.00 ± 1.5	0.13 ± 0.23	0.78 ± 0.14	0.92 ± 1.9	160 ± 200
HpCDFs	5.7 ± 5.0	4.5 ± 1.6	15 ± 0.71	13 ± 27	1700 ± 2000
OCDF	2.8 ± 2.4	2.5 ± 0.90	18 ± 0.71	6.5 ± 12	1400 ± 1500
total PCDFs	36 ± 42	26 ± 11	140 ± 7.1	68 ± 140	5800 ± 5900
DL-PCB congeners[b]					
3,3′,4,4′-TeCB(#77)	7.7 ± 5.1	17 ± 12	450 ± 21	34 ± 55	19 000 ± 32 000
3,4,4′,5-TeCB(#81)	0.64 ± 0.43	0.92 ± 0.76	19 ± 7.1	2.6 ± 5.8	850 ± 1500
2,3,3′,4,4′-PeCB(#105)	58 ± 92	65 ± 49	3800 ± 0.00	110 ± 130	28 000 ± 42 000
2,3,4,4′,5-PeCB(#114)	4.4 ± 6.9	5.3 ± 4.2	210 ± 0.00	7.3 ± 10	2200 ± 3300
2,3′,4,4′,5-PeCB(#118)	140 ± 230	160 ± 130	6900 ± 210	200 ± 210	54 000 ± 78 000
2′,3,4,4′,5-PeCB(#123)	4.5 ± 9	4.3 ± 3.5	170 ± 0.00	6.7 ± 7.5	2000 ± 3000
3,3′,4,4′,5-PeCB(#126)	1.1 ± 1.0	0.58 ± 0.51	41 ± 1.4	2.5 ± 4.7	270 ± 370
2,3,3′,4,4′,5-HxCB(#156)	20 ± 28	7.2 ± 3.5	710 ± 42	29 ± 25	6000 ± 8700
2,3,3′,4,4′,5′-HxCB(#157)	4.6 ± 6.5	1.5 ± 0.54	170 ± 7.1	8.0 ± 7.3	1300 ± 1900
2,3′,4,4′,5,5′-HxCB(#167)	5.7 ± 6.9	3.0 ± 1.5	230 ± 0.00	12 ± 10	2100 ± 3000
3,3′,4,4′,5,5′-HxCB(#169)	1.1 ± 1.2	N.D.[c]	N.D.[c]	0.89 ± 2.0	21 ± 17
2,3,3′,4,4′,5,5′-HpCB(#189)	4.0 ± 8.1	0.26 ± 0.44	21 ± 1.4	2.7 ± 4.2	510 ± 830
WHO-TEQ (ng kg^{-1} dw)	2.7 ± 1.7	2.9 ± 2.4	9.6 ± 0.35	2.3 ± 4.7	190 ± 150

[a] Calculation of SDs was statistically meaningless; however, the values were shown in the same way as other regions.
[b] The IUPAC No. of each DL-PCB congener is provided in parentheses.
[c] Not deteted.

sources were 53% and 30%, respectively. The average ratios of the two congeners in 12 sediment samples from Can Gio and Hue were 54 ± 5% and 28 ± 3%, respectively (a sediment sample from location 5 was excluded from the calculation because no 1,2,3,6,7,8-HxCDD was detected). Therefore, PCDD/PCDFs in the sediment samples from Can Gio and Hue might be attributable to natural sources. At location 7, the concentrations of each of Te, Pe, Hx, Hp and OCDFs was 100, 35, 8.0, 1.2, and 1.0 ng kg^{-1} dw, respectively. These results are consistent with atmospheric deposition originating from various combustion sources (Bakoglu et al., 2005). Given that a town is situated near location 7, pyrogenic sources may have arisen in association with human activities.

In Hanoi, PCDF concentrations were not low (Table 2), and the ratios of each of 1,2,3,7,8,9-HxCDD and 1,2,3,6,7,8-HxCDD to the total 2,3,7,8-substituted HxCDDs were 40 ± 1% and 40 ± 3%, respectively. These values are not consistent with their derivation from natural sources. Therefore, we cannot explain the high occurrence of OCDD in Hanoi. Another characteristic of the PCDD/PCDF profiles obtained for Hanoi is their higher content of PCDF homologues compared with the profiles obtained for Can Gio and Hue. The PCDD/PCDF profiles are also characterized by a reduction in PCDF concentrations with increasing chlorination, as seen at location 7, suggesting that the sedimentary PCDFs in Hanoi are also attributable to various combustion sources (Bakoglu et al., 2005). UNEP (2002) reported that most solid wastes are disposed of at open landfill sites, whereas most medical wastes are disposed of within incinerators. Our data suggest that a possible combustion source in Hanoi is the incineration of medical waste.

A high occurrence of OCDD was also observed in suburban areas of Osaka, as in Can Gio and Hue (Table 2). However, low or undetectable concentrations of PCDFs and specific 2,3,7,8-substituted HxCDD distributions in the areas were not in agreement with those in Can Gio and Hue. Masunaga et al. (2001a,b) reported that a large amount of PCP has been used in the paddy fields of Japans. This suggests that the predominant OCDD in PCDD/PCDFs is attributable to PCP. The dominant homologue except for OCDD was

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TeCDD. The main TeCDD congeners were 1,3,6,8- and 1,3,7,9-TeCDDs, and the average ratios of the two congeners to the total TeCDD congeners (∑TeCDDs) at six suburban locations was >80%. These observations are in agreement with the findings for CNP (Masunaga et al., 2001a).

Another characteristic of PCDD/PCDFs in suburban areas of Osaka is their higher PCDF concentrations compared with those at Can Gio and Hue. The PCDF profiles were characterized by high levels of TeCDFs and HpCDFs. The high abundance of low-chlorinated PCDFs (e.g., TeCDFs) is attributed to various combustion sources, as discussed above (Bakoglu et al., 2005). A high abundance of highly chlorinated PCDFs, such as HpCDFs, is also observed in incinerator-related materials, such as ash and fly ash (Yasuhara et al., 1987). Some municipal and industrial solid-waste incinerators are located in the basin of the Kanzaki River. Until the early 2000s, solid wastes in Japan were also disposed of at small incinerators at schools and in households (MOE, Government of Japan, 2006b). Thus, the incineration of solid wastes is another main source of PCDD/PCDFs in suburban areas of Osaka.

The PCDD/PCDF profiles obtained for urban locations in Osaka are different from those obtained for suburban areas. The profiles are characterized by an abundance of highly chlorinated DD/Fs. Except for the highest peak of OCDD, the profiles are consistent with those of incinerator-related samples (Yasuhara et al., 1987). Several ISWIs are located in the area upstream from locations 23 and 28. The DOEAF (Osaka Prefectural Government) performed a detailed survey of the distribution of PCDD/PCDFs and DL-PCBs in sediment samples around the ISWIs in 2006. The ∑PCDD/PCDF concentrations in 10 sediment samples around ISWIs ranged from 300 000 to 3 000 000 ng kg^{-1} dw (Examination Committee for Environmental Dioxin Pollution [ECFEDP], 2007).

In the present study, we compared the PCDD/PCDF profiles at locations 22, 23, 28, and 29, where extremely high PCDD/PCDF concentrations were detected, with those around ISWIs (ECFEDP, 2007), as shown in Table 3. At locations 23 and 28, a predominance of Hx–OCDD/Fs was observed, with OCDD ≈ Hx–OCDFs > Hx–HpCDDs. With the exception of OCDD, similar characteristics were also observed for samples taken from around ISWIs. The PCDD/PCDF characteristics at the two locations and around the ISWIs

Table 3
Average concentrations of PCDD/PCDF homologues and contribution ratios of both 2,3,7,8-substituted congeners and DL-PCBs to WHO-TEQs in sediment samples from locations 22, 23, 28, and 29, and around ISWIs.

Congeners	Locations				ISWIs[a]
	No. 22	No. 23	No. 28	No. 29	
Concentration (ng kg^{-1} dw)					
TeCDDs	1400	920	890	870	20 000 ± 20 000
PeCDDs	560	1100	650	600	38 000 ± 38 000
HxCDDs	670	2300	1300	860	110 000 ± 100 000
HpCDDs	1500	2300	1800	1200	110 000 ± 88 000
OCDD	5900	3900	3500	3500	110 000 ± 83 000
TeCDFs	730	880	730	750	33 000 ± 21 000
PeCDFs	620	2000	1400	860	72 000 ± 46 000
HxCDFs	720	3800	2600	900	190 000 ± 130 000
HpCDFs	600	4800	3500	830	210 000 ± 150 000
OCDF	340	3600	2500	1500	190 000 ± 130 000
Contribution ratio (%)					
2,3,7,8-substituted PCDDs					
2,3,7,8-TeCDD	11.0	1.9	1.6	2.5	1.5 ± 0.3
1,2,3,7,8-PeCDD	6.3	9.7	11.0	13.0	9.0 ± 1.0
1,2,3,4,7,8-HxCDD	1.1	1.9	2.0	1.3	2.2 ± 0.2
1,2,3,6,7,8-HxCDD	2.4	3.2	3.4	3.6	3.8 ± 0.2
1,2,3,7,8,9-HxCDD	1.5	2.6	2.8	2.8	2.6 ± 0.3
1,2,3,4,6,7,8-HpCDD	3.2	3.0	3.4	2.5	3.3 ± 0.3
OCDD	0.3	0.1	0.1	0.1	0.1 ± 0.0
2,3,7,8-substituted PCDFs					
2,3,7,8-TeCDF	1.6	0.7	1.2	1.6	0.8 ± 0.2
1,2,3,7,8-PeCDF	1.1	1.6	1.6	1.9	1.2 ± 0.1
2,3,4,7,8-PeCDF	10.0	23.0	25.0	12.0	20.5 ± 1.2
1,2,3,4,7,8-HxCDF	3.0	9.2	8.1	4.2	9.9 ± 0.3
1,2,3,6,7,8-HxCDF	2.3	9.5	8.5	3.0	11.0 ± 0.5
1,2,3,7,8,9-HxCDF	0.8	1.1	0.9	0.2	0.5 ± 0.2
2,3,4,6,7,8-HxCDF	3.2	21.0	21.0	3.8	25.9 ± 1.7
1,2,3,4,6,7,8-HpCDF	1.3	6.5	6.9	1.7	7.0 ± 0.8
1,2,3,4,7,8,9-HpCDF	0.2	1.3	1.3	0.2	1.2 ± 0.1
OCDF	0.0	0.1	0.1	0.1	0.1 ± 0.0
DL-PCB congeners[b]					
3,3′,4,4′-TeCB(#77)	3.3	0.1	0.0	1.2	0.0 ± 0.0
3,4,4′,5-TeCB(#81)	0.2	0.0	0.0	0.1	0.0 ± 0.0
2,3,3′,4,4′-PeCB(#105)	3.8	0.1	0.0	2.8	0.0 ± 0.0
2,3,4,4′,5-PeCB(#114)	1.4	0.0	0.0	1.2	0.0 ± 0.0
2,3′,4,4′,5-PeCB(#118)	6.7	0.4	0.1	5.8	0.0 ± 0.0
2′,3,4,4′,5-PeCB(#123)	0.2	0.0	0.0	0.2	0.0 ± 0.0
3,3′,4,4′,5-PeCB(#126)	33.0	2.0	1.8	27.0	0.7 ± 0.1
2,3,3′,4,4′,5-HxCB(#156)	2.9	0.2	0.1	3.8	0.0 ± 0.0
2,3,3′,4,4′,5′-HxCB(#157)	0.7	0.1	0.0	0.9	0.0 ± 0.0
2,3′,4,4′,5,5′-HxCB(#167)	0.0	0.0	0.0	0.0	0.0 ± 0.0
3,3′,4,4′,5,5′-HxCB(#169)	0.1	0.1	0.1	0.1	0.1 ± 0.0
2,3,3′,4,4′,5,5′-HpCB(#189)	0.0	0.0	0.0	0.1	0.0 ± 0.0

[a] ECFEDP, 2007.
[b] The IUPAC No. of each DL-PCB congener is provided in parentheses.

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agree well with those of incinerator-related samples (Yasuhara et al., 1987). Therefore, the PCDD/PCDF profiles at locations 23 and 28 mainly reflect the influence of ISWIs. The higher content of OCDD at the two locations is also associated with PCP (Yasuhara et al., 1987). At locations 22 and 29, OCDD and ∑PCDFs dominantly occurred in PCDD/PCDFs, as also observed at locations 23 and 28. However, their profiles were not consistent with those around ISWIs. Therefore, PCDD/PCDFs at the two locations 23 and 28 were possibly derived from PCP and other types of incineration, rather than from ISWIs.

3.3. DL-PCB concentrations in sediment samples from Vietnam and Japan, and determination of their main emission sources

Few studies have focused on environmental DL-PCB pollution in Vietnam. As shown in Table 2, the ∑DL-PCB concentrations in Can Gio were the same as those in Hue and suburban locations in Osaka, but lower than those in urban regions of Hanoi and Osaka. The average concentration in Hanoi was approximately 40 times higher than that in Can Gio, in contrast to ∑PCDD/PCDF concentrations. At urban locations in Osaka, extremely high ∑DL-PCB concentrations were observed at locations 22 and 29 (370 000 and 300 000 ng kg^{-1} dw, respectively). Concentrations of DL-PCB in Can Gio, Hue, and suburban locations in Osaka were the same as those in suburban locations in Tokyo, whereas those in Hanoi and urban areas of Osaka were as high as those in urban regions of Tokyo.

The main source of DL-PCBs was estimated using the ratio of the sum of 3,3′,4,4′,5-PeCB (#126) and 3,3′,4,4′,5,5′-HxCB (#169) to the sum of 3,3′,4,4′-TeCB (#77), 3,3′,4,4′,5-PeCB (#126), and 3,3′,4,4′,5,5′-HxCB (#169). The proportion of DL-PCBs in commercial PCBs was approximately 1% (Kannan et al., 1987), whereas that from combustion sources was approximately 50% (Sakai et al., 1994). The value of (#126 + #169)/(#77 + #126 + #169) in Can Gio ranged from 13% to 50%, with an average of 19 ± 15%, suggesting that the DL-PCBs in Can Gio are attributable to both commercial PCBs and pyrogenic sources. The commercial PCBs would result from the use of imported electrical products (Kishida et al., 2007). Other possible sources of commercial PCBs are military activities during the Second Indochina War and the Vietnam War (Thao et al., 1993), High proportions of pyrogenic DL-PCBs were found at locations 3, 4, 6, and 7 (19–50%). There exists a town near location 3 and a hotel near locations 6 and 7; furthermore, a shrimp nursery is situated close to location 4. Therefore, these pyrogenic DL-PCBs may be primarily derived from human activities. Artillery fire and exploding bombs during the wars are another possible combustion source (Thao et al., 1993). The DL-PCB concentrations in Can Gio were very low.

The average value of (#126 + #169)/(#77 + #126 + #169) was 3 ± 3% in Hue and 8 ± 0% in Hanoi. These values are similar to those of commercial PCBs, although a large difference in DL-PCB concentrations was observed between the two regions. This finding possible arose because commercial PCBs and imported electrical products containing PCBs are mainly used in urban regions such as Hanoi and HCMC (Kishida et al., 2007).

In Osaka, the average value of (#126 + #169)/(#77 + #126 + #169) was 9 ± 5% at suburban locations and 6 ± 4% at urban locations. These values correspond to those for commercial PCBs. The use and production of commercial PCBs in Japan have been prohibited by law since 1973. Therefore, DL-PCBs in the sediment samples from Osaka would have originated from the greater use of commercial PCBs in urban areas before the 1970s (Kishida et al., 2007). Since the 1970s, Japanese law has required that commercial PCBs are adequately managed. However, extremely high DL-PCB concentrations were detected locally at locations 22 and 29, although the concentration at location 23 (22 000 ng kg^{-1} dw), located only a few kilometers upstream from location 22, was less than 10% of the concentration at location 22. These observations may be attributable to the outflow of commercial PCBs into the environment as a result of their inadequate management.

3.4. WHO-TEQ values in sediment samples from Vietnam and Japan

Table 2 also lists the WHO-TEQ values for sediment samples from five regions of Vietnam and Japan. The WHO-TEQ values in the sediment samples from Can Gio, derived from 29 congeners of 2,3,7,8-substituted PCDD/PCDFs and DL-PCBs, ranged from 0.42 to 7.1 ng kg^{-1} dw, with an average value of 2.7 ± 1.7 ng kg^{-1} dw. The values in Can Gio were the same as those in Hue, Hanoi, and suburban areas of Osaka, but much lower than those in urban locations of Osaka. Among the eight urban locations in Osaka, high WHO-TEQ values were obtained at four locations: 22 (240 ng kg^{-1} dw), 23 (370 ng kg^{-1} dw), 28 (260 ng kg^{-1} dw), and 29 (260 ng kg^{-1} dw).

Compared with values obtained for other Vietnamese and Japanese regions, WHO-TEQ values in Can Gio, etc. were as high as those in areas of the Aluoi Valley aerially sprayed with Agent Orange (0.6–17 ng kg^{-1} dw; Dwernychuk et al., 2002), suburban areas in Tokyo (0.2–2.4 ng kg^{-1} dw; BOE, Tokyo Metropolitan Government, 2009), and Akita (0.021–5.3 ng kg^{-1} dw; Kiguchi et al., 2007), whereas values obtained at urban locations in Osaka were as high as those around former storage depots for herbicides in the Aluoi Valley (2.0–901.22 ng kg^{-1} dw; Dwernychuk et al., 2002) and Bien Hoa (4.6–437.6 ng kg^{-1} dw; Mai et al., 2007), and at urban locations in Tokyo (maximum: 290 ng kg^{-1} dw; BOE, Tokyo Metropolitan Government, 2009).

3.5. Contribution of each source of PCDD/PCDFs and DL-PCBs to WHO-TEQ values in Vietnam and Japan

We now consider the dominant contribution of each source of PCDD/PCDFs and DL-PCBs to WHO-TEQ values in the sediments from Vietnam and Osaka. The average proportional contribution of each of the 29 congeners to the WHO-TEQ values can be obtained from Table 2. In Can Gio, the average contribution ratio of TCDD to WHO-TEQ was 31 ± 11%, being the highest among the 29 congeners. Previous studies have shown that high contribution ratios of TCDD are attributable to Agent Orange (Dwernychuk et al., 2002; Mai et al., 2007). Dwernychuk et al. (2002) reported that the contribution ratio of TCDD to I-TEQ in the soil and sediment samples from the Aloui Valley ranged from 78% to >99% at former storage depots for Agent Orange and from 41% to 90% at areas aerially sprayed with Agent Orange. In the present study, values in Can Gio were lower than those in the Aloui Valley. However, the average contribution ratio in Can Gio was higher than those in Hue, Hanoi, and suburban and urban locations in Osaka. These findings suggest only minor residual concentrations of sedimentary TCDD caused by aerial spraying with Agent Orange in Can Gio.

In Can Gio and Hue, 2,3,7,8-substituted PCDD congeners, except for TCDD (e.g., 1,2,3,7,8-PeCDD, 1,2,3,7,8,9-HxCDD, 1,2,3,4,6,7,8-HpCDD, and 1,2,3,6,7,8-HxCDD) are the main contributors to WHO-TEQ values. The contribution ratios of the sum of 1,2,3,7,8-PeCDD, 1,2,3,7,8,9-HxCDD, 1,2,3,4,6,7,8-HpCDD, and 1,2,3,6,7,8-HxCDD in Can Gio and Hue are approximately 40% and 60%, respectively, higher than the values for TCDD. Therefore, aerial spraying with Agent Orange was not a main contributor in either region. Gadomski et al. (2004) and Gaus et al. (2001) reported that the higher contribution ratios of 1,2,3,7,8-PeCDD, 1,2,3,7,8,9-HxCDD, 1,2,3,4,6,7,8-HpCDD, and 1,2,3,6,7,8-HxCDD are attributable to natural origins. Therefore, the dominant contributor to WHO-TEQ values in Can Gio was possibly natural sources rather than aerial spraying with Agent Orange, as in the other rural region (Hue).

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In contrast, in the urban region of Hanoi, congeners of #126 and 2,3,4,7,8-PeCDF were the main contributors to the WHO-TEQ values (43% and 13%, respectively). As discussed above, DL-PCBs are mainly derived from the past use of commercial PCBs. The congener 2,3,4,7,8-PeCDF, which has the highest contribution ratio among the 2,3,7,8-substituted PCDD/PCDFs, is attributable to pyrogenic sources (Yasuhara et al., 1987), mainly the incineration of medical waste in Hanoi. Therefore, in Hanoi, the main contribution to WHO-TEQ values is commercial PCBs, possibly followed by the incineration of medical waste.

In Osaka, 2,3,7,8-substituted PCDD/PCDF and DL-PCB congeners contributed similarly to WHO-TEQ values at suburban locations. The dominant congeners at suburban locations were 1,2,3,7,8-PeCDD, #126, 2,3,4,7,8-PeCDF, 1,2,3,4,6,7,8-HpCDD, and TCDD. High contributions by 1,2,3,7,8-PeCDD and 1,2,3,4,6,7,8-HpCDD were observed in CNP and PCP, respectively (Masunaga et al., 2001a). As mentioned above, the high contribution of combustion-related 2,3,4,7,8-PeCDF is attributable to the incineration of solid waste, and the main origins of DL-PCBs are the commercial PCBs produced and used in the past. Therefore, at suburban locations in Osaka, the herbicides used for rice cultivation, the incineration of solid wastes, and commercial PCBs that had been produced and used until the 1970s contributed similarly to the obtained WHO-TEQ values. However, very low WHO-TEQ values were recorded in suburban areas.

At urban locations in Osaka, the contribution ratio of 2,3,7,8-substituted PCDFs to WHO-TEQ was larger than those of 2,3,7,8-substituted PCDDs and DL-PCBs. The dominant 2,3,7,8-substituted PCDF congeners at urban locations were 2,3,4,7,8-PeCDF, 2,3,4,6,7,8-HxCDF, 1,2,3,6,7,8-HxCDF, and 1,2,3,4,7,8-HxCDF. The contribution ratio of 2,3,4,7,8-PeCDF was the highest among the 2,3,7,8-substituted PCDD/PCDFs and DL-PCBs. The average contribution ratio of the total 2,3,7,8-substituted PCDFs to WHO-TEQ (51%) was higher than that at suburban locations (31%). These data indicate that the contribution of the incineration of solid wastes to WHO-TEQ values could be higher at urban locations than at suburban locations.

To estimate the causes of the significantly increased WHO-TEQ values at locations 22, 23, 28, and 29, we compared the profiles at the four locations with those around ISWIs (ECFEDP, 2007), as shown in Table 3. The profiles at locations 23 and 28 are similar to those around the ISWIs, with the dominant congeners being 2,3,4,7,8-TeCDF and 2,3,4,6,7,8-HxCDFs. The contribution ratios of the total 2,3,7,8-substituted PCDFs to WHO-TEQ at the two locations were approximately 75%, with these values being as high as those around the ISWIs (average: 81 ± 3%). These data indicate that the increased WHO-TEQ values at areas downstream from IWSIs (e.g., locations 23 and 28) are strongly associated with incinerator-related materials from the ISWIs.

In contrast, the dominant congeners at locations 22 and 29 were #126 > 2,3,4,7,8-PeCDF ≈ 1,2,3,7,8-PeCDD. This finding suggests that commercial PCBs that might have been inadequately managed made a dominant contribution to WHO-TEQ values at locations 22 and 29, rather than the incineration of solid waste.

In summary, the residual sedimentary TCDD contributed by the aerial spraying of Agent Orange occur in low concentrations in Can Gio. The main contributors to the WHO-TEQ values in Can Gio are natural sources, as in Hue. In contrast, commercial PCBs are the dominant contributors to WHO-TEQ values in Hanoi. In Osaka, PCDD/PCDF and DL-PCB pollution in sediments from suburban locations is caused equally by agrochemicals used in rice cultivation, the incineration of solid waste, and commercial PCBs. The dumping of incinerator-related materials and/or the inadequate management of commercial PCBs have resulted in significantly elevated WHO-TEQ values of 240–370 ng kg^{-1} dw at urban locations.

Acknowledgement

We gratefully thank Mr. M. Kawashima, Osaka University, for providing us with three sediment samples from Can Gio (Nos. 8–10).

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