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Contamination by PCDDs, PCDFs and coplanar PCBs in open dumping sites in Vietnam – levels, patterns and toxic implications

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Abstract Open landfill dumping areas for municipal wastes in Asian developing countries have recently received particular attention with regard to environmental pollution problems. In this study, concentrations of polychlorinated dibenzo-*p*-dioxins (PCDDs), polychlorinated dibenzofurans (PCDFs) and coplanar polychlorinated biphenyls (PCBs) were determined in soils collected from open dumping sites in Vietnam and other Asian countries such as Philippines, Cambodia and India. Residue concentrations of PCDD/Fs and coplanar PCBs in dumping site soils were apparently greater than those in soils collected in agricultural or urban areas far from dumping sites, suggesting that dumping sites are potential sources of PCDD/Fs and related compounds. PCDD/F levels in soils from Vietnamese dumping sites were lower than those in soils from other countries, indicating less dioxin contamination in dumping sites from Vietnam among Asian developing countries surveyed. Homologue profiles of PCDD/Fs in dumping site soils Hochiminh City were similar to those of typical environmental sink samples, while one soils from Hanoi dumping site reflected pattern of samples representing typical emission samples. This result suggests the sporadic formation of dioxins in dumping sites from Hanoi and less extent of recent formation from Hochiminh City. On the other hand, data from other countries like Cambodia, Philippines and India showed clear evidence for the recent formation of dioxins. Uncontrolled combustions of solid wastes by waste pickers, generation of methane gas and low temperature burning can be major factors for the formation of dioxins in dumping sites. Elevated fluxes of PCDD/Fs to soils in dumping sites were encountered in Philippines, Cambodia, India and Hanoi, Vietnam, and these levels were higher than those reported for other countries. Considerable loading rates of PCDD/Fs in the dumping sites of these countries were observed, ranging from 20 to 3900 mg/yr (0.12 – 35 mg TEQ/yr). PCDD/F concentrations in some soil samples from Philippines, Cambodia, India and Hanoi, Vietnam exceeded environmental guideline values, suggesting potential health effects on humans and wildlife living near these dumping sites. To our knowledge, this

is the first comprehensive study on PCDD/Fs contamination in open dumping sites in Vietnam as well as in other Asian developing countries.

Keywords Open dumping sites, PCDD/Fs, uncontrolled burning

Introduction

In recent years, public media have voiced concern regarding the role of the open dumping sites in Asian developing countries, where large amounts of municipal solid wastes have been dumped, as a potential source of toxic contaminant. Unfortunately, in most of the developing countries in Southeast Asia, such open dump areas are located near human habitats, and therefore, exposure to various toxic chemicals originated from dumping sites are of serious concern over the effects on human health, wildlife and environmental quality (Tanabe, 2002). Uncontrolled burning of solid waste by waste pickers and generation of methane gas, lack of advanced waste incineration technology and natural low temperature burning are major problems in dumping sites in Asian developing countries at present. These are favorable factors for the formation of dioxins and dibenzofurans.

Because of these reasons, we anticipated elevated contamination by PCDD/Fs in dumping site environments in Asian developing countries. Our laboratory has conducted a comprehensive research to find an answer to the question whether open dumping sites in Asian developing countries are potential sources of dioxins and related compounds. In this study, soil samples were collected in open dumping sites from Vietnam, Philippines, Cambodia and India and concentrations of PCDDs and related compounds such as PCDFs and coplanar polychlorinated biphenyls (PCBs) were measured. Soils from agricultural and urban areas, located far from dumping sites were also examined for PCDD/Fs to elucidate the differences in dioxin accumulation in dumping sites and agricultural areas and whether dumping sites are sources of PCDD/Fs. The annual loads of dioxins into dumping site areas were also estimated to give further insight into the fate of dioxins in tropical environment.

Material and methods

Samples and sampling locations

Soil samples were collected in two biggest cities in Vietnam, Hanoi and Hochiminh City and in Manila (Philippines), Phnom Penh (Cambodia) and Chennai (India) during 1999-2001. Details of samples and characteristic of dumping sites are given in Table 1. Those open dumping sites are located close to human habitat and there are large numbers of people scavenging in these dumpsites for collecting recycling wastes. In dumping sites in Cambodia, uncontrolled burning of solid wastes was observed. Soil samples were collected at depths from 0 – 10 cm, at 5 points with an area of approximately 25 m², and combined together and considered as a representative sample. Urban or agricultural soils were also collected at some locations far from the dumping site areas (at least 30 km far from dumping sites). Soil samples

collected from these locations hereinafter are referred to as “control sites” relative to dumping sites, from the following discussion. Soils were kept in clean plastic bags, maintained at 4°C, transported to laboratory and stored at -20°C until chemical analysis.

Chemical analysis.

Chemical analyses of PCDD/Fs and coplanar PCBs followed the method described in our recent study (Minh *et al.*, 2003). Recoveries for ¹³C₁₂-labeled PCDD/Fs and coplanar PCBs, which were added prior to multi-layer silica gel column, ranged within 60 – 120 %. Variation of concentrations between duplicate analyses of soil samples was within 15 % for congener concentrations with signal-to-noise (s/n) ratio > 50, and was within 30 % for those with s/n ratio of 5 – 50.

Table 1. Information of soil samples and dumping sites in Asian developing countries

Country (City)	Site names	Collected	Number of samples		Working time (year)	Area (m ²)	Capacity (t/day)	Remark
		year	Dumping sites	Control sites				
Philippines (Quezon City)	Payatas	1999	3	not available	from 1993 till now	230000	200	Low temperature burning observed
Cambodia (Phnom Penh)	Stoeung Meanchey	1999/2000	15	4	from 1979 till now	30000	200	Low temperature and burning by waste pickers
India (Chennai)	Perungudi	2000	13	5	from 1985 till now	1400000	1300	Low temperature burning observed
Vietnam (Hanoi)	Tay Mo	2000	9	1	from 1997 to 1999	50000	1360	Uncontrolled burning observed
Vietnam (Hochiminh)	Dong Thanh	2001	8	3	from 1990 to 2001	300000	4000	Low temperature burning observed

Results and discussion

Residue concentrations

Average concentrations of PCDD/Fs in soils from dumping sites were the highest in Philippines, followed by soils in Cambodia, India, Hanoi and Hochiminh City, Vietnam in order (Table 2). Elevated residues found in soils from dumping sites in Philippines and Cambodia, ranging from 44,000 to 75,000 and 330 to 200,000 pg/g dry wt, respectively. In particular, few soil samples in Cambodia had extremely high concentrations of PCDD/Fs (up to 200,000 pg/g dry wt). Soils from dumping site in Hochiminh City, Vietnam accumulated the lowest PCDD/F residues (ranged from 130-260 pg/g dry wt). Soils collected from agricultural or urban areas located far from dumping sites were considered as the control samples for corresponding dumping soils. Interestingly, the control levels of PCDD/Fs ranged from 33 to 370 pg/g dry wt, which were substantially lower than those in soils from dumping sites in most of the countries surveyed, except in Hochiminh City (Table 2). The magnitude of contamination of dumping site soils was apparently greater than that in control sites, suggesting that open dumping sites are a potential source of dioxins and related compounds in Asian developing countries.

Trend of contamination by non- and mono-*ortho* coplanar PCBs was somewhat similar to that of PCDD/Fs with the concentrations ranging from 900 to 42,000 pg/g dry wt in dumping sites and from 3 to 160 pg/g dry wt in control sites (Table 2). Their contribution to total TEQs was also different between dumping and control site, which were 10–57% and 5-20%, respectively. In dumping sites, the highest coplanar PCBs concentration was found in Philippines and the lowest was in Hochiminh City, Vietnam. However, their concentrations in the control group were the highest in Hochiminh City, followed by Hanoi (North Vietnam), India and Cambodia (control site in Philippines was not investigated). This result indicates considerable background contamination of coplanar PCBs in Vietnam. On the other hand, relatively lower residues of PCDD/Fs in Vietnamese soils indicate less contamination of these compounds in dumping sites as compared to other Asian developing countries in region.

Table 2. Concentrations (pg/g dry wt) of PCDD/Fs and coplanar PCBs in soils from dumpsites and reference sites in Asian countries

Countries	India		Vietnam-Hanoi		Vietnam-Hochiminh		Cambodia		Philippines
	DS (n = 13)	RS (n = 5)	DS (n = 9)	RS (n = 1)	DS (n = 8)	RS (n = 3)	DS (n = 15)	RS (n = 4)	DS (n = 3)
T ₄ CDD	920	0.55	1350	5	9.4	11	5500	12	6300
P ₂ CDD	1000	0.57	590	5.5	11	5.7	6900	12	6700
H ₆ CDD	1300	1.1	510	26	27	18	5300	16	11000
H ₇ CDD	800	3.4	210	56	53	29	1200	11	5800
O ₈ CDD	1900	21	480	270	220	100	1400	39	12000
T ₄ CDF	490	1.1	1400	9.2	13	12	4600	18	8600
P ₂ CDF	330	0.8	850	1	9.5	3.9	2700	10	4900
H ₆ CDF	290	1	490	< 0.06	10	3.7	1900	9.1	3300
H ₇ CDF	230	1.4	160	0.6	9.4	2.1	520	3.4	1800
O ₈ CDF	110	1.9	38	< 0.1	4.9	1.2	140	1.4	500
Total PCDD/Fs	7400	33	6100	370	370	190	30000	130	61000
Range	2 200 - 34 000	18 - 79	125 - 50 500	370	21 - 880	130 - 260	330 - 200 000	40 - 370	44 000 - 75 000
Total co-PCBs	6670	28	2600	130	910	160	7600	11	41500
Range	1300 - 20 000	12 - 52	59 - 7100	120	65 - 2000	46 - 3000	590 - 24 000	1.1 - 27	11 000 - 83 000
TEQs^a	52	0.22	102	1.1	2.7	1.3	395	30	540

^aTEQs were calculated using WHO-TEF (Van den Berg et al., 1998)

Residue levels of PCDD/Fs from the soils of dumping sites were compared with those in soils from some other locations over the world that were reported as dioxin-contaminated sites (Figure 1). The cited data from literature represent the concentrations in soils from the environments that are impacted from the municipal waste incinerator, open landfill dumping sites and HCH-manufacturing waste sites (Minh *et al.*, 2003). Interestingly, concentrations of PCDD/Fs in some soil samples from Cambodia, Philippines and India were higher than those reported in soils from Ohio, USA (mean concentration of 15700 pg/g dry wt), which was near a municipal solid waste incinerator emitting large amount of dioxins (1000 g TEQ/yr). Similarly, an open landfill site in Crete, Greece, where uncontrolled low temperature combustion was noticed for more than 10 years, has an extremely high concentration of PCDD/Fs (92000 pg/g dry wt) in a site close to the burning areas (Martens *et al.*, 1998). Likewise, three soil samples collected in Cambodia, two in Philippines and one in Hanoi, Vietnam contained PCDD/F concentrations > 50,000 pg/g dry wt.

Given that there were no industrial or waste incineration activities in all those areas, elevated concentrations of PCDD/Fs again indicate that open dumping sites with uncontrolled burning processes are significant sources of dioxin formation. In this context, however, further studies to examine the extent of PCDD/Fs contamination along the gradient in the vicinity of the dumping sites would provide more detailed insights into the impacts of dumping sites emissions.

Homologue profiles

To further understand the role of the dumping sites as a source of PCDD/Fs, we examined the homologue profiles of PCDD/Fs in dumping soils and control soils. Their PCDD/Fs homologue profiles were then compared with typical profiles of samples representing environmental sources (municipal waste incinerator emissions) and sinks (urban soils, sediments or atmospheric deposition samples) (Figure 2).

Samples representing environmental source were an emission of a typical municipal waste incinerator in the United States (Brzuzny and Hites, 1996) and an average of 12 different combustion sources (Baker and Hites, 2000), while environmental sinks were soils collected from various locations over the world by Hites and co-workers in an Indiana University laboratory (Baker and Hites, 2000). In general, the homologue profiles of sample representing environmental sources are characterized by the predominance of lower chlorinated dibenzofurans and an increasing proportion from tetra- to hexachlorinated dibenzo-*p*-dioxins.

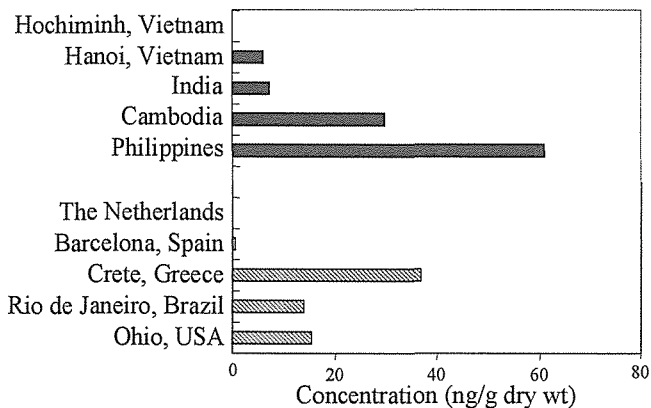


Figure 1. Comparison of PCDD/F residues in soils from open dumping sites in Asian countries with those in soils from dioxin-contaminated areas in the world. See Minh et al., (2003) for further information of cited data from dioxin-contaminated areas.

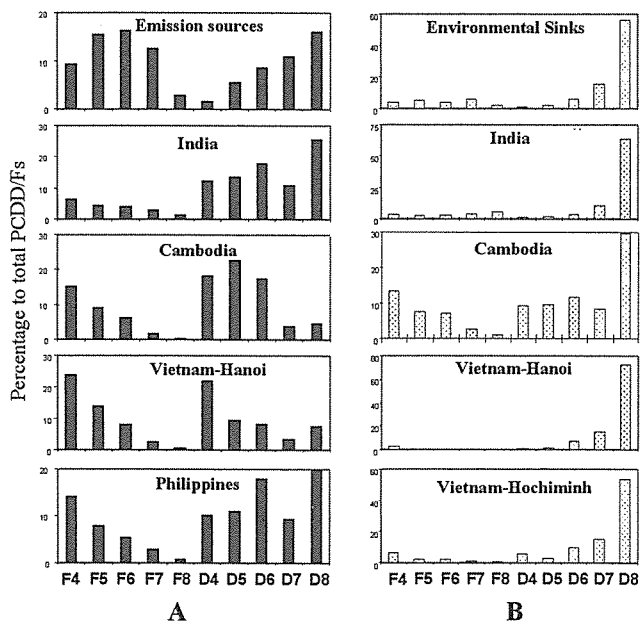


Figure 2. Comparison of homologue profiles of PCDD/Fs in soils from Asian dumping sites with those in samples representing emission sources (A) and environmental sinks (B)

On the other hand, the typical pattern of environmental sink samples contains O₈CDD as predominant congener.

Comparisons of PCDD/Fs homologue pattern of samples in the investigated areas with these typical patterns of sources and sinks could provide insight into the sources and status of formation of PCDD/Fs in dumping sites. Interestingly, homologue profiles of the dumping soils from Philippines, India, Cambodia and one soil sample from Hanoi, Vietnam reflect pattern of emission sources. Profiles of the control soils, in contrast, were similar to those of typical environmental sinks (Figure 2). PCDD/F profiles of dumping soils from Philippines and Cambodia were most similar to those of emission sources, suggesting recent formation of dioxins in these dumping sites. Homolog profiles in soils from Hochiminh City (Vietnam) resemble those of environmental sink samples and also similar to those of soils from control sites in other countries (Figure 2). This fact and together with relatively low residue levels of PCDD/Fs and coplanar PCBs might indicate less dioxin formation in dumping sites in Hochiminh City.

Flux and load of PCDD/Fs to the dumping sites

In this study, we used the approach that was reported by Hites and co-workers (Brzuzy and Hites, 1995, 1996; Wagrowski and Hites, 2000) to estimate the flux of PCDD/Fs to the soils and their load to the dumping site areas in Asian developing countries. The details of the calculation of flux to soils of PCDDs and PCDFs and their loading rate were described in our original paper (Minh *et al.*, 2003).

Table 3. Estimated fluxes of PCDD/Fs to soils in dumping sites from Asian developing countries in comparison with those to soils from other countries in the world.

Country	Flux (ng/m ² /yr)		References
	Mean	Range	
Philippines	17000	13000 - 21000	Present study
Cambodia	2900	31 - 19000	Present study
India	990	290 - 4500	Present study
Vietnam-Hanoi	4100	83 - 34000	Present study
Vietnam-Hochiminh	67	3.8 - 160	Present study
Hong Kong	2800	840 - 4030	Wagrowski and Hites (2000)
Australia	13	10 - 16	Wagrowski and Hites (2000)
Lake Baikal, Russia	9	not available	Wagrowski and Hites (2000)
Thailand	2	not reported	Wagrowski and Hites (2000)
Indiana, USA	1280	1260 - 1290	Wagrowski and Hites (2000)
Michigan, USA	80	52 - 110	Wagrowski and Hites (2000)
British Columbia, Canada	64	4.2 - 350	Wagrowski and Hites (2000)
Yukon Territory, Canada	6.6	1.2 - 15	Wagrowski and Hites (2000)
Brazil	2.2	not available	Wagrowski and Hites (2000)
UK	180	170 - 220	Wagrowski and Hites (2000)
Spain	100	72 - 130	Wagrowski and Hites (2000)
Germany	97	51 - 140	Wagrowski and Hites (2000)
Norway	26	not available	Wagrowski and Hites (2000)

Estimated fluxes of PCDD/Fs to soils in dumping sites in Philippines, Cambodia, India and Vietnam in comparison with agricultural/urban soils from other countries are given in Table 3. It is interesting to note that fluxes to dumping site soils from Philippines and Cambodia were greater than those from other locations in the world, including some contaminated sites in Hong Kong and Indiana, USA (Table 3). This result clearly supports the fact that dumping sites are potential sources of PCDD/Fs. Fluxes to control soils were apparently lower than those to dumping site soils. Considering the homologue pattern of soils from control sites, it can be suggested that soils from control sites were mainly originated from atmospheric depositions. Elevated fluxes observed in dumping sites in the present study could be attributed to uncontrolled combustion processes in a large open landfill area.

Table 4. Estimated load of PCDD/Fs to dumping site areas in Asian developing countries.

Country	Dumping site area (m ²)	Load	
		mg/yr	mg TEQ/yr
Philippines	230000	3900	35
Cambodia	30000	87	1.1
India	1400000	1400	8.8
Vietnam-Hanoi	50000	210	3.2
Vietnam-Hochiminh	300000	20	0.12

The load of PCDD/Fs to the dumping sites indicate that dumping sites in Philippines and India with a huge area of approximately 23 and 140 ha, could receive the highest annual amount of 3900 and 1400 mg/yr PCDD/Fs (35 and 8.8 mg TEQs/yr), respectively (Table 4). Dumping site in Hochiminh City, Vietnam, had the lowest loading rate due to the less contamination of PCDD/Fs in soils. As for comparison, total annual fluxes to the Kanto region, Japan, one of the most polluted areas in the world, were estimated and was found to range from 50 to 900g TEQ with a total area of 32,000 km² (approximately 3 millions ha) (Ogura *et al.*, 2001). The area of dumping sites in India is 140 ha, which is 21,000 times smaller than that of Kanto region; and this area was estimated to receive an amount of 8.8 mg TEQs every year. These estimation data suggest that dumping sites in India and Philippines may be a significant reservoir for PCDD/Fs. Possible impacts on human health and wildlife living near dumping sites are of great concern and deserve further comprehensive studies.

Risk assessment

The formation of PCDD/Fs in open dumping sites in Asian developing countries has raised a considerable public concern to human health for not only communities living near the dumping sites but also for people who live far away because PCDD/Fs may undergo atmospheric transport and deposit in such distant areas. For risk assessment of soils

contaminated by dioxins and related compounds, the Agency for Toxic Substances and Disease Registry (ATSDR) proposed guidelines recommending that areas having soil concentrations within the range from 50 to 1000 pg TEQ/g should be evaluated for bioavailability, ingestion rates, community concerns, etc., and soils with the concentrations over 1000 pg TEQs/g should be considered for stronger actions like health studies, exposure investigations, etc. (ATSDR, 1997).

Japanese government recently issued new standards for dioxins in soil, which established level of 1000 pg TEQ/g for maximum acceptable level and those within 250 – 1000 pg TEQ/g must be kept under surveillance (Japan's new dioxin standard, 1999). Results of this study revealed that many soil samples in dumping sites contained TEQ concentrations exceeding 250 pg/g TEQs (Figure 3), suggesting the necessity of continuous monitoring. Particularly, some soils from Cambodia and Hanoi dumping sites contained TEQ concentrations beyond the level of 1000 pg/g, suggesting their potential for causing adverse health risk for humans and wildlife.

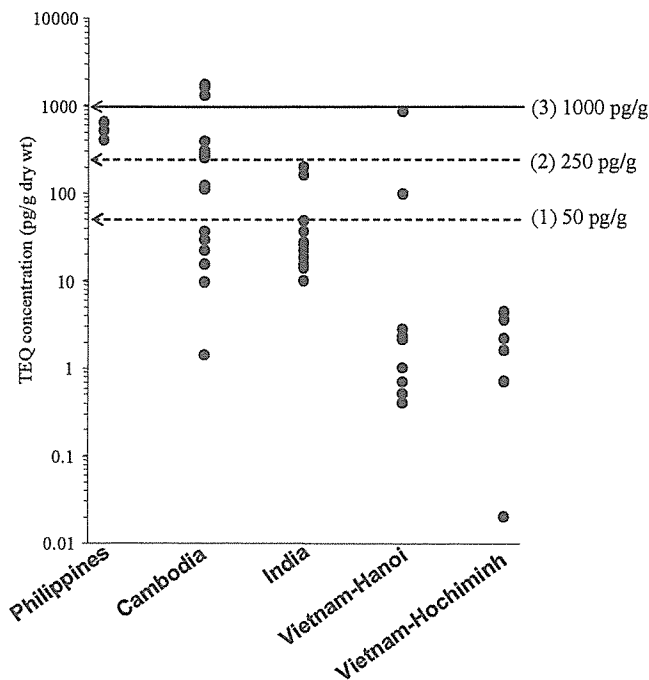


Figure 3. PCDD/F concentrations (pg/g dry wt) in soils from dumping sites in Asian developing countries in comparison with various environmental guideline values. (1) 50 pg/d TEQs dry wt, recommended by ATSDR for bioavailability, ingestion rate, etc.; (2) 250 pg/g TEQs dry wt, recommended by Japanese Government for the continuous monitoring necessity; (3) 1000 pg/g TEQs dry wt, Japanese Standard and level proposed by ATSDR for stronger action and research on exposure and health studies.

Conclusions and recommendations for future research

To our knowledge, this is the first comprehensive study on PCDD/Fs pollution in open dumping sites in Asian developing countries. Our result provides clear evidence that dumping sites in Asian developing countries are potential sources of PCDD/Fs and dioxin-like PCBs. Based on the results of this study; we have addressed a new environmental issue that open landfill dumping areas in developing countries may be a “hot spot” of pollution by toxic chemicals. Thus, dioxin contamination may become a key environmental problem in Asian developing countries in the 21st century. Possible toxic effects on human health and wildlife living near dumping areas should be studied in future and this is a critical step for further considerations of the necessary control measures towards protecting environmental quality.

Monitoring studies on exposure to other chemicals such as organochlorine insecticides, which were used in large quantities in the past and have been used until very recently in Asian developing countries, are indispensable. Temporal trends of contamination of PCDD/Fs and organochlorine insecticides also deserve comprehensive investigations.

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