Contributions of dry and wet depositions of polychlorinated dibenzo-*p*-dioxins and dibenzofurans to a contaminated site resulting from a penetachlorophenol manufacturing process

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Abstract The soils at a factory for manufacturing pentachlorophenol were heavily contaminated by polychlorinated dibenzo-*p*-dioxins and dibenzofurans (PCDD/Fs). In order to verify the contributions of dry and wet deposition of PCDD/Fs from the ambient air, the concentration of PCDD/Fs in ambient air and soil were measured, the partition of particle- and gas-phases of atmospheric PCDD/Fs was calculated, and the annual fluxes of total dry and wet PCDD/F depositions were modeled. Average atmospheric PCDD/F concentration was 1.24 ng Nm^{-3} (or 0.0397 ng I-TEQ Nm^{-3}). Moreover, over 92.8% of total PCDD/Fs were in the particle phase, and the dominant species were high chlorinated congeners. The total PCDD/F fluxes of dry and wet deposition were 119.5 ng m^{-2} $year^{-1}$ (1.34 ng I-TEQ m^{-2} $year^{-1}$) and 82.0 ng m^{-2} $year^{-1}$ (1.07 ng I-TEQ m^{-2} $year^{-1}$), respectively. By scenario simulation, the total fluxes of dry and wet PCDD/F depositions were 87.1 and 68.6 ng I-TEQ, respectively. However, the estimated PCDD/F contents in the contaminated soil were 839.9 µg I-TEQ. Hence, the contributions of total depositions of atmospheric PCDD/F were only 0.02%. The results indicated that the major sources of PCDD/F for the contaminated soil could be attributed to the pentachlorophenol manufacturing process.

Keywords PCDD/Fs · Partition ·
Pentachlorophenol · Flux · Deposition

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Introduction

Polychlorinated dibenzo-*p*-dioxins and polychlorinated dibenzofurans (PCDD/Fs) have received considerable attention recently, because those congeners are harmful to human health (USEPA

2000). Combustion is thought to be the major source of PCDD/F to the atmosphere, and in Taiwan, the dominant sources are iron sintering, coal-fired power plants, and electric arc furnaces, which contribute 32%, 28%, and 23% to the total inventory (USEPA 2000; Rappe 1992; Lin et al. 2007; Shih et al. 2009).

PCDD/F congeners can be transported over long distances in the air and thus be deposited anywhere by either dry or wet deposition (Van Jaarsveld and Schutter 1993; Tysklind et al. 1993). Dry deposition, including particle and gas phases, includes aerodynamic transport, boundary layer transport, and interaction with the receptor (Lee et al. 1996a). Previous research indicated that the dry deposition flux of PCDD/F increases as the temperature decreases and also that the removal efficiency of PCDD/F by dry deposition increases with increasing levels of chlorination (Koester and Hites 1992). The other process is wet deposition, with precipitation as the removal mechanism (Lohmann and Jones 1998). Rain scavenging is most efficient for particle-bound PCDD/Fs and for PCDD/Fs with higher levels of chlorination, and one study reported that wet deposition is the major pathway for highly chlorinated congeners to the bare soil (Koester and Hites 1992; Schröder et al. 1997). In addition, the fluxes of PCDD/F deposition in the rainy season have been shown to be higher than those in the dry season (Kaupp and McLachlan 1999).

In Southern Taiwan, there is one extremely contaminated site, which is polluted by pentachlorophenol (PCP), mercury, and PCDD/Fs. This is An-Shun Site of the China Petrochemical Development Corporation (ASS-CPDC) which was formerly a chloralkali and PCP factory, and the related chemical processes produced sodium hydroxide, PCP, and other chlorinated derivatives, such as PCDD/Fs (Alcock and Jones 1997; Kauppinen et al. 1993; Hagenmaier and Brummer 1987; USEPA 1994; EPBTC 2004, 2009). This is because chlorine was used to chlorinate the non-substituted phenol to produce chlorophenols, which are important precursors for the formation of PCDDs (Wang et al. 2003a, b). A similar previous study showed that soil samples were measured and the concentrations were as high as 0.239–1357 ng I-TEQ g^{-1} (Soong et al. 1997).

In order to verify the major source of PCDD/Fs contributions for the soil at the An-Shun site, this study investigates the atmospheric PCDD/F concentration in this contaminated site and particularly its concentration of particle- and gas-phase atmospheric PCDD/F. Further, the annual dry and wet depositions of PCDD/Fs on the contaminated site were calculated with the use of a model. In addition, the contribution of PCDD/F from ambient air to the contaminated soil was also estimated.

Methods

Sampling

Five ambient PCDD/F samples were collected near/in the contaminated area of the ASS-CPDC. The sampling locations were decided according to the assessment of the Industrial Source Short-Term model for modeling the PCDD/F concentration in the region (Yang et al. 1998; Mi et al. 2001; Lee et al. 2003). The PCDD/F concentrations in the ambient air were collected by a standard semi-volatile sampling train (General Metal Works PS-1) based on the revised US EPA Reference Method TO9A (USEPA 1999). Prior to sampling, XAD-2 resin was spiked with PCDD/F surrogate standards prelabeled with isotopes (Wang et al. 2003a, b). A total of 10 samples were collected in summer and winter. In addition, three soil samples were collected from the tussock in the ASS-CPDC. The sampling method was based on the National Environment Analysis Method of Taiwan (NIEA SS102.60B).

Analysis of PCDD/FS

The analyses of the ambient air samples followed the US EPA Reference Method TO9A (USEPA 1999). Samples were extracted with toluene for 24 h, and this was then followed by a series of sample cleanup procedures. The extract was transferred to a vial and finally further concentrated by a N_2 gas stream.

A high-resolution gas chromatograph mass spectrometer (HRGC/MS) was used for PCDD/F analysis. The HRGC/MS (Hewlett–Packard 6970

Series Gas, CA) was equipped with a DB-5 fused silica capillary column (L = 60 m, ID = 0.25 mm, film thickness = 0.25 μm; J&W Scientific, CA) with a splitless injection, while the HRMS (Micromass Autospec Ultima, Manchester, UK) had a positive electron impact source. The analyzer mode of the selected ion monitoring was used with the resolving power set at 100.00. The electron energy and source temperature were specified at 35 eV and 250°C, respectively. The oven temperature program was set as follows: initially at 150°C (held for 1 min), then increased by 30°C min^{-1} to 220°C (held for 12 min), and finally increased by 1.5°C min^{-1} to 310°C (held for 20 min). Helium was used as the carrier gas. The protocol for quality analysis/quality control was strictly followed (Wang and Lee 2010).

Particle/gas phase partition and dry/wet deposition of PCDD/FS

Particle and gas concentration was calculated by gas/particle partitioning multiplying total PCDD/Fs concentrations. Gas/particle partitioning was calculated by partitioning constant as follows (Eq. 1; Yamasaki et al. 1982; Pankow 1994; Chao et al. 2004; Hung et al. 2002):

$$K_p = \frac{F/\text{TSP}}{A} \qquad (1)$$

Where F (pg m^{-3}) is the particle-phase PCDD/F concentration, A (pg m^{-3}) is the gas-phase PCDD/F concentration, K_p (m^3 μg^{-1}) is a partitioning constant, and TSP (μg m^{-3}) is the concentration of total suspended particulates (Pankow 1991, 1994; Pankow and Bidleman 1992; Yamasaki et al. 1982).

K_p was calculated by Eqs. 2 and 3.

$$\log K_p = mr \times \log \text{PL}° + br \qquad (2)$$

$$\log \text{PL}° = -1.34\,(\text{RI})/T + 1.67 \times 10^{-3}\,(\text{RI}) \\ - 1320/T + 8.087 \qquad (3)$$

Plotting log K_p against the logarithm of the subcooled liquid vapor pressure, where PL° is the subcooled liquid vapor pressure, RI is the gas chromatographic retention indexes, and T is ambient temperature (K) (Eitzer and Hites 1988;

Hung et al. 2002; Donnelly et al. 1987; Hale et al. 1985; Chao et al. 2004). In the previous study, Chao did find subcooled PL°-based model to be excellent descriptors for the gas–particle partitioning of PCDD/Fs (Chao et al. 2004). This model led to the well estimation of C_p, C_g and increased the reliability of calculation for PCDD/F deposition fluxes.

The dry deposition of PCDD/Fs in the atmosphere could have two forms, particle and gas phase, which are calculated by Eq. 4.

$$F_T = F_g + F_p, \qquad (4)$$

Where F_T is the total fluxes of PCDD/F deposition, F_g is the gas phase fluxes and F_p is the particle phase fluxes.

The average dry deposition velocity of total PCDD/Fs in Taiwan has been measured and found to be 0.42 cm s^{-1} (Shih et al. 2006; Sheu et al. 1996; Lee et al. 1996a, b). However, the dry deposition velocity of gas-phase PCDD/Fs is one of the uncertainties for lacking the real measurement data. A value of 0.010 cm s^{-1} was proposed for gas-phase polycyclic aromatic hydrocarbon dry deposition velocity (Sheu and Lee 1996) and was used in this study because PCDD/Fs and PAHs were all SVOC.

The wet deposition of PCDD/Fs is associated with vapor dissolution into rain and the removal of suspended particulate by precipitation.

S_g is the gas scavenging ratio of PCDD/Fs, which can be defined by Eq. 5 and calculated by Eq. 6.

$$S_g = RT/H \qquad (5)$$

$$S_g = C_{\text{rain,dis.}}/C_g \qquad (6)$$

Where R is the universal gas constant (82.06× 10^{-6} m^3 atm mol^{-1} K^{-1}), T is the ambient temperature (K), and H is the Henry constant (m^3 atm mol^{-1}). $C_{\text{rain,dis}}$ is the dissolved phase of PCDD/F concentration in the rain, and C_g is the gas phase of PCDD/F concentrations in ambient air.

The S_p is the particle scavenging ratio of PCDD/Fs, which can be calculated by Eq. 7.

$$S_p = C_{\text{rain,particle}}/C_p \qquad (7)$$

Where $C_{rain,particle}$ is the particle phase of PCDD/F concentrations in the rain, and C_p is the particle phase of the PCDD/F concentrations in ambient air.

The S_{tot} is the total scavenging ratio of PCDD/Fs, which can be calculated by Eq. 8.

$$S_{tot} = S_g (1 - \Phi) + Sp \times \Phi \qquad (8)$$

Where Φ is the fraction of the total air concentration bound to particles.

The particle scavenging ratios of PCDD/Fs and the S_p values of OCDD and OCDF are based on Eitzer and Hites (1989).

Results and discussion

PCDD/F contents in the contaminated and background soils

The PCDD/F contents in the contaminated and background soil are shown in Table 1. The conge-ner profiles of PCDD/Fs in the contaminated and background soil are shown in Fig. 1a and b, respectively. The PCDD/F contents in the contaminated soil ranged from 9,735 to 35,970 ng I-TEQ kg^{-1}, with a mean PCDD/F content of 4,307 ng I-TEQ kg^{-1} (1,934,450 ng kg^{-1}; EPBTC 2004; Lee et al. 2008). In Brazil, the soil sample collected from a contaminated site in a hexachlorocyclohexane plant was as high as 13,900 ng I-TEQ kg^{-1} (Braga et al. 2002).

The extremely high PCDD/F contents in the contaminated soil were over 6,500 times higher than that of the background soil (0.658 ng I-TEQ kg^{-1}) and clearly exceeding the regulated soil pollution standard in Taiwan (1,000 ng I-TEQ kg^{-1}; EPA Taiwan 2009). That is, the chemical factory produced not only sodium hydroxide and pentachlorophenol products but also other chlorinated derivatives, such as PCDD/Fs (Hagenmaier and Brummer 1987; Kauppinen et al. 1993; USEPA 1994; Alcock and Jones 1997; EPBTC 2004). Figure 1a shows the dominant

Table 1 PCDD/F contents in contaminated soil and background soil

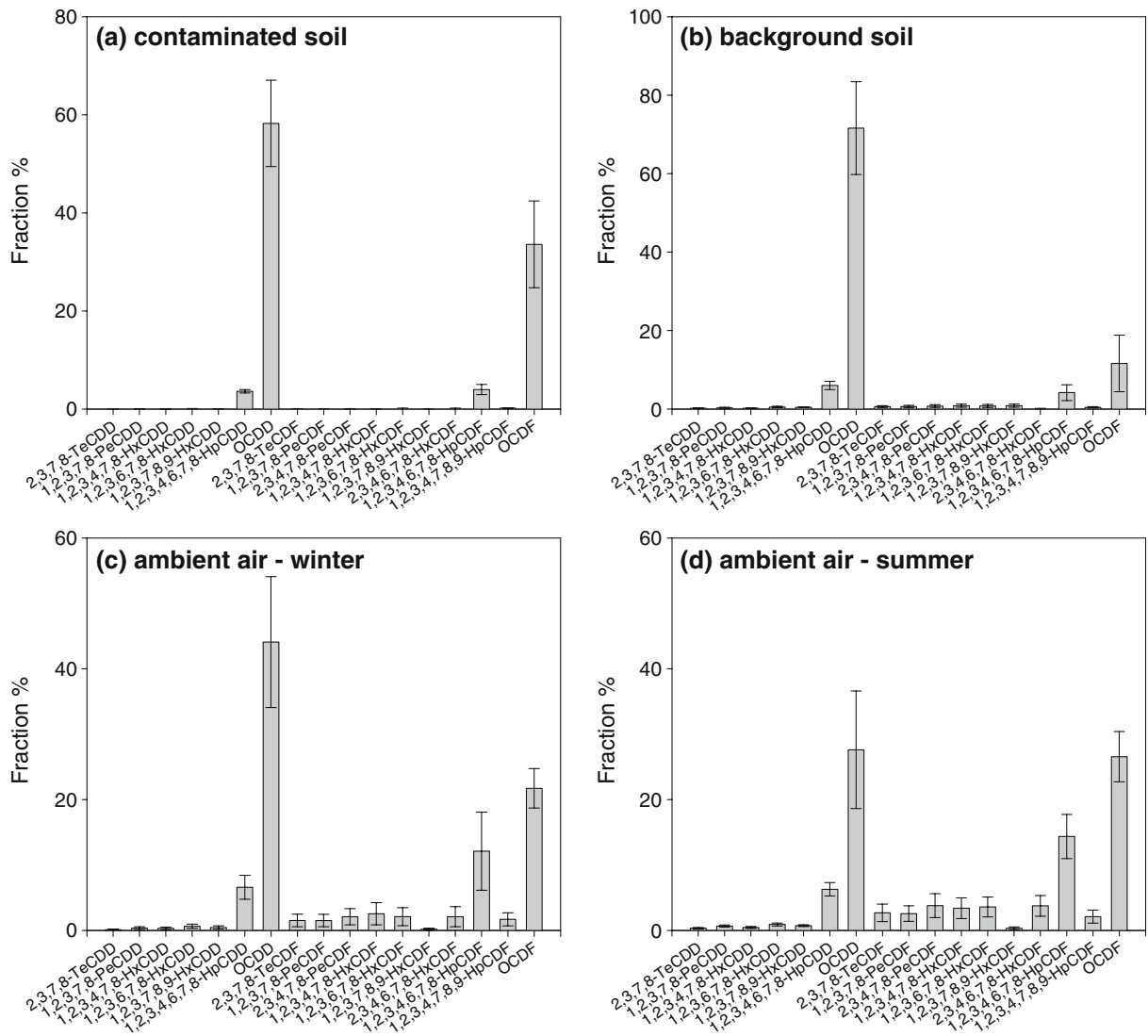| PCDD/Fs	Contaminated soil, mean ($n = 3$)	RSD (%)	Background soil, mean ($n = 5$)	RSD (%)
2,3,7,8-TeCDD	29.2	46.9	0.088	62.8
1,2,3,7,8-PeCDD	102	22.4	0.144	45.3
1,2,3,4,7,8-HxCDD	351	58.3	0.0895	30.0
1,2,3,6,7,8-HxCDD	1,659	45.0	0.243	34.3
1,2,3,7,8,9-HxCDD	487	14.7	0.220	28.2
1,2,3,4,6,7,8-HpCDD	70,028	47.5	2.90	42.8
OCDD	1,126,612	53.7	34.6	43.8
2,3,7,8-TeCDF	559	66.5	0.281	35.3
1,2,3,7,8-PeCDF	326	62.8	0.298	48.9
2,3,4,7,8-PeCDF	504	35.2	0.366	46.5
1,2,3,4,7,8-HxCDF	764	9.0	0.413	43.6
1,2,3,6,7,8-HxCDF	1,241	62.7	0.373	47.1
1,2,3,7,8,9-HxCDF	142	59.6	0.413	38.7
2,3,4,6,7,8-HxCDF	1,551	31.2	0.0487	51.1
1,2,3,4,6,7,8-HpCDF	76,897	45.9	2.02	37.7
1,2,3,4,7,8,9-HpCDF	3,734	39.3	0.203	27.5
OCDF	649,464	64.6	5.61	66.8
PCDDs	1,199,267	53.2	38.2	42.6
PCDFs	735,182	61.8	10.0	54.1
PCDDs/PCDFs ratio	1.63	–	3.82	–
Total PCDD/Fs (ng kg^{-1})	1,934,450	51.6	48.3	32.3
PCDDs ng I-TEQ kg^{-1}	2,157	46.4	0.279	43.4
PCDFs ng I-TEQ kg^{-1}	2,150	34.9	0.379	42.9
PCDDs/PCDFs(TEQ) ratio	1.00	–	0.737	–
Total PCDD/F I-TEQ (ng I-TEQ kg^{-1})	4,307	36.6	0.658	37.1

Fig. 1 Congener profiles of PCDD/F **a** contaminated soil; **b** background soil; **c** ambient air in the winter; **d** ambient air in the summer

congeners in the polluted soil were OCDD and OCDF.

PCDD/F concentration in the ambient air

The PCDD/F concentrations in the atmosphere during two seasons (winter, summer) are shown in Table 2. The samples were sampled in summer and winter because the wind directions were mainly two directions in Taiwan and were typically distributed in summer and winter. The means

of atmospheric PCDD/F concentration in winter and summer were 1.95 pg Nm^{-3} (0.0514 pg I-TEQ Nm^{-3}) and 0.531 pg Nm^{-3} (0.0279 pg I-TEQ Nm^{-3}), respectively, lower than the regulated value in Japan (0.6 pg I-TEQ Nm^{-3}; MOE Japan 2008). The annual mean of PCDD/F concentration was 1.24 pg Nm^{-3}, with a I-TEQ value of 0.0397 pg I-TEQ Nm^{-3}, which is slightly lower than the other areas in Taiwan (0.088 pg I-TEQ Nm^{-3}; Wang et al. 2005). A previous study also reported that the PCDD/F concentration in winter

Table 2 Atmospheric PCDD/F concentrations in winter and summer

PCDD/Fs	Winter, mean ($n = 5$)	RSD (%)	Summer, mean ($n = 5$)	RSD (%)	Average
2,3,7,8-TeCDD	0.00206	29.8	0.00203	19.6	0.00205
1,2,3,7,8-PeCDD	0.00671	15.6	0.00402	23.2	0.00536
1,2,3,4,7,8-HxCDD	0.00588	23.7	0.00273	18.1	0.00431
1,2,3,6,7,8-HxCDD	0.0122	16.4	0.00558	21.3	0.00888
1,2,3,7,8,9-HxCDD	0.00815	39.0	0.00443	28.0	0.00629
1,2,3,4,6,7,8-HpCDD	0.128	27.9	0.0389	35.7	0.0835
OCDD	0.858	61.5	0.171	35.7	0.515
2,3,7,8-TeCDF	0.0294	42.3	0.0166	29.7	0.0230
1,2,3,7,8-PeCDF	0.0292	46.3	0.0159	25.3	0.0226
2,3,4,7,8-PeCDF	0.0406	42.3	0.0234	28.3	0.0320
1,2,3,4,7,8-HxCDF	0.0494	48.6	0.0210	27.3	0.0352
1,2,3,6,7,8-HxCDF	0.0408	47.9	0.0221	26.0	0.0314
1,2,3,7,8,9-HxCDF	0.00378	38.2	0.00191	15.9	0.00285
2,3,4,6,7,8-HxCDF	0.0406	50.5	0.0232	26.3	0.0319
1,2,3,4,6,7,8-HpCDF	0.236	11.9	0.0889	28.2	0.162
1,2,3,4,7,8,9-HpCDF	0.0328	28.3	0.0130	30.1	0.0229
OCDF	0.423	24.1	0.164	50.2	0.293
PCDDs	1.02	54.1	0.229	83.6	0.625
PCDFs	0.925	19.4	0.302	29.7	0.614
PCDDs/PCDFs ratio	1.10	–	0.756	–	1.02
Total PCDD/Fs (pg Nm^{-3})	1.95	28.1	0.531	49.4	1.24
PCDDs pg I-TEQ Nm^{-3}	0.0102	34.2	0.00588	21.9	0.00802
PCDFs pg I-TEQ Nm^{-3}	0.0412	46.5	0.0221	26.6	0.0317
PCDDs/PCDFs(TEQ) ratio	0.247	–	0.266	–	0.254
Total PCDD/Fs I-TEQ (pg I-TEQ Nm^{-3})	0.0514	43.9	0.0279	20.9	0.0397

was four to eight times higher than in summer in rural Germany due to the demand for domestic heating; and atmospheric PCDD/F concentration also varies with the seasons, because of photolysis and its effects on chemical reactions (Hippelein et al. 1996; Lohmann and Jones 1998).

Figure 1c, d shows the congener profiles of the 17 2,3,7,8-substituted PCDD/Fs in winter and summer. The dominant congeners in the ambient air are highly chlorinated congeners, such as OCDD, OCDF, 1,2,3,4,6,7,8-HpCDF, and 1,2,3,4,6,7,8-HpCDD.

The particle- and gas-phase concentrations of the PCDD/Fs are listed in Table 3, and the related fractions are shown in Fig. 2a (winter) and b (summer). The particle- and gas-phase PCDD/F concentrations were 1.81 and 0.140 ng Nm^{-3}, respectively, in winter, while 0.372 and 0.247 ng Nm^{-3} in summer. The mean partitions of particle and gas phases were 92.8% and 7.18%, respectively, in winter, but were 60% and 40% in summer. Hence,

in ambient air, the most of the PCDD/Fs were thus mainly in the particle phase, as well as a higher particle-phase PCDD/F concentration in winter than in summer. The difference of mean partitions in the particle phase for in winter and summer should be caused by the various pollution sources transferred from the different wind direction, that is, in different seasons (summer and winter) in Taiwan.

Dry and wet deposition of PCDD/FS

The fluxes of dry and wet PCDD/F deposition are listed in Table 4. The results show that the fluxes of dry deposition in particle and gas phases were 119 ng m^{-2} year^{-1} (1.28 ng I-TEQ m^{-2} year^{-1}) and 0.484 ng m^{-2} year^{-1} (0.0629 ng I-TEQ m^{-2} year^{-1}), respectively. Hence, particle-bound deposition contributed 99.6% of dry deposition flux of PCDD/F. The total fluxes of dry PCDD/F deposition were

Table 3 Atmospheric PCDD/F concentrations in particle (Cp) and gas phases (Cg) and in winter and summer	PCDD/Fs	Winter, Cp	Winter, Cg	Summer, Cp	Summer, Cg
	2,3,7,8-TeCDD	0.000296	0.00176	0.0000216	0.00201
	1,2,3,7,8-PeCDD	0.00330	0.00341	0.000210	0.00381
	1,2,3,4,7,8-HxCDD	0.00494	0.000944	0.000578	0.00215
	1,2,3,6,7,8-HxCDD	0.0103	0.00186	0.00123	0.00435
	1,2,3,7,8,9-HxCDD	0.00703	0.00112	0.00107	0.00336
	1,2,3,4,6,7,8-HpCDD	0.124	0.00372	0.0235	0.0154
	OCDD	0.854	0.00439	0.152	0.0192
	2,3,7,8-TeCDF	0.00292	0.0265	0.000120	0.0165
	1,2,3,7,8-PeCDF	0.00948	0.0197	0.000445	0.0155
	2,3,4,7,8-PeCDF	0.0163	0.0243	0.00088	0.0225
	1,2,3,4,7,8-HxCDF	0.0363	0.0131	0.00271	0.0183
	1,2,3,6,7,8-HxCDF	0.0304	0.0104	0.00297	0.0191
	1,2,3,7,8,9-HxCDF	0.00313	0.000648	0.0003813	0.001529
	2,3,4,6,7,8-HxCDF	0.0324	0.00824	0.00395	0.0193
	1,2,3,4,6,7,8-HpCDF	0.221	0.0152	0.0365	0.0524
	1,2,3,4,7,8,9-HpCDF	0.0318	0.00102	0.00764	0.00536
	OCDF	0.420	0.00331	0.1379	0.0261
	PCDDs	1.00	0.0172	0.178	0.0503
	PCDFs	0.803	0.122	0.194	0.196
	PCDDs/PCDFs ratio	1.25	0.141	0.92	0.256
	Total PCDD/Fs (pg Nm^{-3})	1.81	0.140	0.372	0.247
	PCDDs pg I-TEQ Nm^{-3}	0.00627	0.00390	0.000802	0.00507
	PCDFs pg I-TEQ Nm^{-3}	0.0221	0.0192	0.00206	0.0201
	PCDDs/PCDFs (TEQ) ratio	0.284	0.203	0.390	0.252
	Total PCDD/Fs I-TEQ (pg I-TEQ Nm^{-3})	0.0283	0.0231	0.00286	0.0252

119.5 ng m^{-2} year^{-1} (1.34 ng I-TEQ m^{-2} year^{-1}). In Bloomington, the average fluxes of dry deposition PCDD/F were 160 ng m^{-2} year^{-1} (Koester and Hites 1992).

The wet deposition fluxes of PCDD/F in particle and dissolved phases were 80.5 ng m^{-2} year^{-1} (0.869 ng I-TEQ m^{-2} year^{-1}) and 1.48 ng m^{-2} year^{-1} (0.196 ng I-TEQ m^{-2} year^{-1}), respectively, with a total wet PCDD/F deposition flux of 82.0 ng m^{-2} year^{-1} (1.07 ng I-TEQ m^{-2} year^{-1}; Table 4). Similar to the dry deposition flux, particulate-phase deposition contributed 98.2% of wet PCDD/F deposition flux.

The results show that the particle bound deposition contributed more PCDD/Fs than gas phase for dry deposition or dissolved phase for wet deposition, the dry deposition (119.5 ng m^{-2} year^{-1}) contributed more PCDD/Fs than wet deposition (82.0 ng m^{-2} year^{-1}) in this site, and the total deposition flux of PCDD/F was 201 ng m^{-2} year^{-1} (2.41 ng I-TEQ m^{-2} year^{-1}). Hence, the more significant mechanism of PCDD/F deposition was from the particle bound deposition of dry deposition. In addition, a positive relationship between total deposition flux and rainfall was reported (Wallenhorst et al. 1997), meaning that the total deposition fluxes of PCDD/F are affected by meteorological conditions.

Estimation of PCDD/F deposition from ambient air to the contaminated soil

The pentachlorophenol factory studies in this work occupied 150,000 m^2 and operated from 1942 to 1982. This scenario was simulated for 1 × 1 m area for a 65-year period to estimate the total PCDD/Fs deposition (including dry and wet deposition) from ambient air to the surface of the soil. Annual meteorological conditions from 2000 to 2007 are listed in Table 5. Average meteorological parameters were used to estimate the contributions of PCDD/Fs from ambient air to the contaminated soil.

The modeling results show that the total dry and wet depositions of PCDD/Fs were 7,743.5 ng (87.1 ng I-TEQ) and 5,291.7 ng (68.6 ng I-TEQ), respectively. However, total PCDD/Fs in the con- taminated soil was as high as 377,217 μg (839.9 μg I-TEQ). Hence, the atmospheric PCDD/F de- position contributed only 0.02% (by I-TEQ). In duck farms, the contribution of PCDD/F from

Fig. 2 Distribution of total PCDD/Fs in gas (*G*) and particle (*P*) phases: **a** in the winter; **b** in the summer
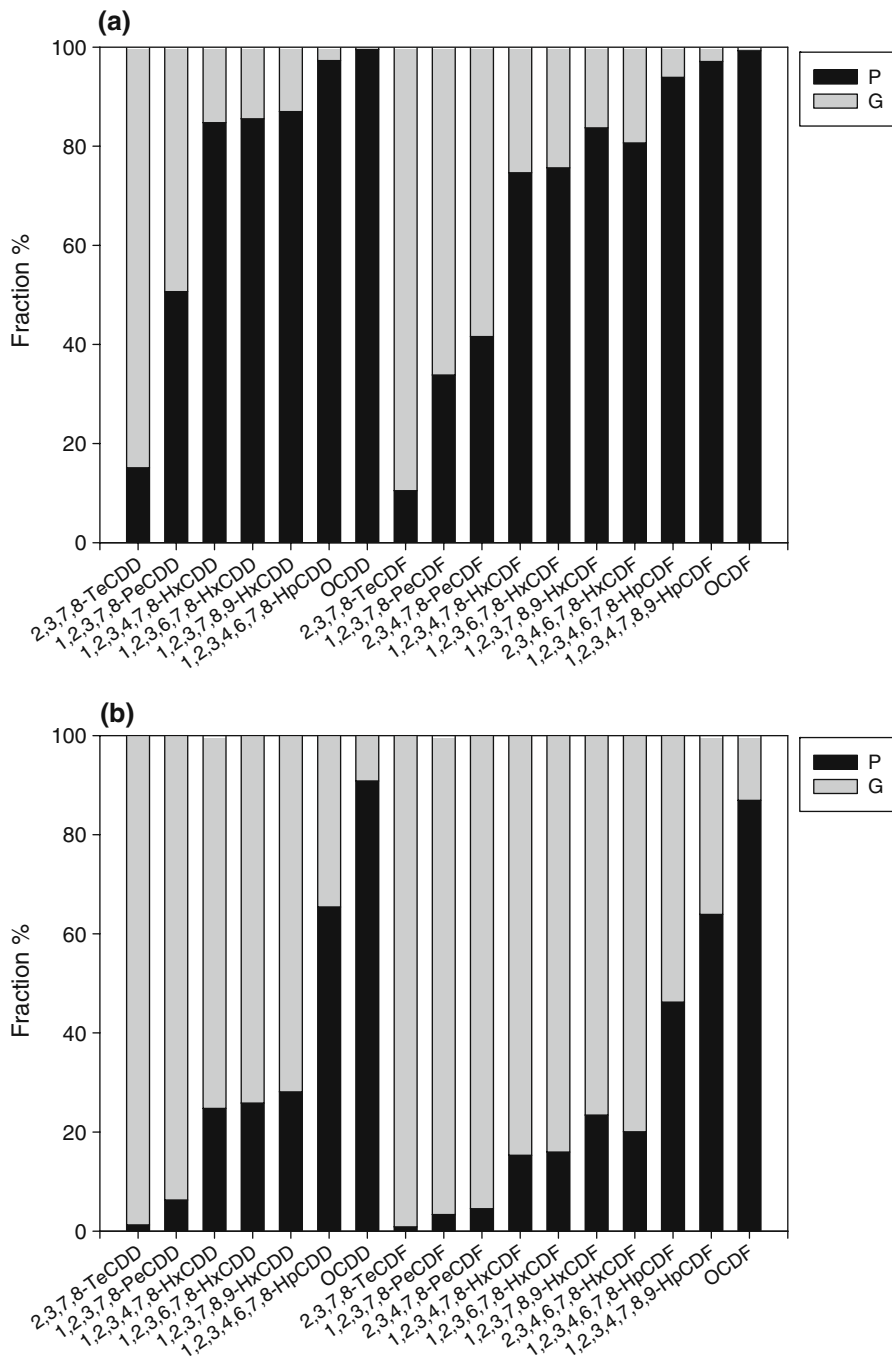
Table 4 Dry and wet deposition fluxes of PCDD/F

PCDD/Fs	Fd,g	Fd,p	Fd	Dissolved	Particle	Fw
2,3,7,8-TeCDD	0.00438	0.00963	0.0140	0.00262	0.00653	0.00915
1,2,3,7,8-PeCDD	0.00965	0.115	0.124	0.0728	0.0779	0.151
1,2,3,4,7,8-HxCDD	0.00434	0.263	0.267	0.00192	0.178	0.180
1,2,3,6,7,8-HxCDD	0.00866	0.557	0.565	0.00382	0.377	0.381
1,2,3,7,8,9-HxCDD	0.00573	0.414	0.420	0.00253	0.281	0.284
1,2,3,4,6,7,8-HpCDD	0.0226	8.18	8.20	0.0355	5.55	5.58
OCDD	0.0284	56.0	56.0	0.0831	38.0	38.1
2,3,7,8-TeCDF	0.0498	0.0733	0.123	0.0664	0.0497	0.116
1,2,3,7,8-PeCDF	0.0449	0.273	0.318	0.178	0.185	0.363
2,3,4,7,8-PeCDF	0.0611	0.511	0.572	0.242	0.346	0.588
1,2,3,4,7,8-HxCDF	0.0474	1.55	1.60	0.0652	1.05	1.12
1,2,3,6,7,8-HxCDF	0.0414	1.43	1.47	0.0570	0.97	1.03
1,2,3,7,8,9-HxCDF	0.00299	0.168	0.171	0.00412	0.114	0.118
2,3,4,6,7,8-HxCDF	0.0371	1.70	1.74	0.0510	1.15	1.21
1,2,3,4,6,7,8-HpCDF	0.0853	13.8	13.9	0.120	9.35	9.47
1,2,3,4,7,8,9-HpCDF	0.00657	2.22	2.23	0.00921	1.51	1.52
OCDF	0.0241	31.5	31.5	0.481	21.3	21.8
PCDDs	0.0838	65.5	65.6	0.202	44.4	44.6
PCDFs	0.401	53.2	53.6	1.27	36.1	37.3
PCDDs/PCDFs ratio	0.209	1.23	1.44	0.159	1.23	1.20
Total PCDD/Fs (ng m^{-2} year^{-1})	0.484	119	119.5	1.48	80.5	82.0
PCDDs (ng I-TEQ m^{-2} year^{-1})	0.0113	0.328	0.340	0.0403	0.223	0.263
PCDFs (ng I-TEQ m^{-2} year^{-1})	0.0516	0.953	1.01	0.156	0.647	0.803
PCDDs/PCDFs (TEQ) ratio	0.220	0.344	0.564	0.258	0.344	0.328
Total PCDD/Fs I-TEQ (ng I-TEQ m^{-2} year^{-1})	0.0629	1.28	1.34	0.196	0.869	1.07

ambient air to soil was estimated to be 0.242–0.611% (Lee et al. 2009). Therefore, the major source of PCDD/Fs for the contaminated soil can be assumed to be the pentachlorophenol production process.

Table 5 Meteorological conditions from 2000 to 2007

Time	Temperature (°C)	Rainfall (mm)	PM$_{10}$ (μg m^{-3})
2007	24.94	2,207	73.7
2006	24.98	1,867	75.5
2005	24.57	3,149	76.4
2004	24.61	1,108	73.6
2003	24.81	899	65.2
2002	24.86	1,212	62.5
2001	24.45	2,107	65.9
2000	24.37	1,754	70.5
Average	24.70	1,788	70.4

Conclusions

The mean atmospheric PCDD/F concentration in winter and summer was 0.0514 and 0.0279 pg I-TEQ Nm^{-3}, respectively, with an annual average PCDD/F concentration of 0.0397 pg I-TEQ Nm^{-3}, which is slightly lower than other areas in Taiwan. In ambient air, the PCDD/F contributions of particle- and gas-bound deposition were 92.8% and 7.18%, respectively, in winter, as well as 60% and 40%, respectively, in summer. For dry/wet depositions, PCDD/Fs were also mainly contributed from the particulate-phase deposition, reaching 99.6% for total dry deposition flux and 98.2% for wet deposition flux. Moreover, dry deposition flux of PCDD/F (1.34 ng I-TEQ m^{-2} year^{-1}) was higher than wet deposition (1.07 ng I-TEQ m^{-2} year^{-1}).

According to the scenario simulation, the total flux of dry and wet PCDD/F deposition was about 13.04 μg, far lower than the estimated PCDD/F contents in the contaminated soil (377,217 μg). The contribution of total depositions of atmospheric PCDD/F was only 0.02% (by I-TEQ). The results show that the major source of PCDD/F for the contaminated soil was due to inappropriate disposal, the PCDD/F containing wastes were mixed with or dump in the soil and resulted in so high content of PCDD/Fs in the soil.

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