Dry and Wet Deposition of Polychlorinated Dibenzo-*p*-dioxins and Dibenzofurans on the Drinking Water Treatment Plant

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ABSTRACT

This study investigated the concentrations and congener profiles of polychlorinated dibenzo-*p*-dioxins (PCDDs) and polychlorinated dibenzofurans (PCDFs) in intake source water (source water) and tap drinking water (tap water) of drinking water treatment plants (DWTPs). In order to have a better understanding on the influence of atmospheric PCDD/F deposition on drinking water, PCDD/Fs in the ambient air of a DWTP (DWTP-LN) were measured and both dry and wet deposition on the water treatment facilities were assessed. The results of this study indicated that the mean PCDD/F concentration in tap water (0.0039 pg WHO-TEQ/L) was found to be approximately 55% of magnitude less than that in source water (0.0086 pg WHO-TEQ/L). In addition, the total deposition flux (dry + wet) of PCDD/Fs entering the DWTP-LN was 27.0 ng I-TEQ/m^2-year, and wet and dry deposition contributed approximately 12.0% and 88%, respectively. It reveals that dry deposition is more important than wet deposition of PCDD/Fs in the ambient air of DWTP-LN. Atmospheric deposition of PCDD/Fs will increase the level in source water of DWTP-LN up to 8.91×10^{-3} pg I-TEQ/L, which is approximately 92% of the PCDD/Fs in source water. If a removal efficiency of 87% is achieved by conventional treatment processes including coagulation, flocculation, sedimentation and rapid sand filtration, the water after treatment may increase 1.16×10^{-3} pg I-TEQ/L, which is approximately 43% of the concentration level in tap water. These results indicate that in the DWTP-LN, the influence of atmospheric deposition of PCDD/Fs on the drinking water is of great significance, and water treatment facility with a cover is suggested.

Keywords: Dioxins; Source water; Tap water; Dry deposition; Wet deposition.

INTRODUCTION

Due to the potential adverse health effects from exposure to polychlorinated dibenzo-*p*-dioxins (PCDDs) and polychlorinated dibenzofurans (PCDFs), such as reproductive difficulties and increased risk of cancer (US EPA, 2003), their presence in the environment is of great concern. In addition, PCDD/Fs are persistent in the environment, remaining intact for long periods (Stockholm Convention, 2001). Furthermore, PCDD/Fs are typically hydrophobic and lipophilic, thus in aquatic systems and soils they partition strongly to solids rather than the aqueous phase (Jones and de Voogt, 1999). Because of these properties, they become widely distributed, accumulate in the fatty tissue of living organisms and are toxic to humans and wildlife (Stockholm Convention, 2001).

In Japan, the environmental quality standard for dioxins in water is 1 pg WHO-TEQ/L or less (Environmental Health Department, 2002). While in the USA, the highest level of 2,3,7,8-TCDD that is allowed in drinking water is 3×10^{-8} mg/L (equal to 30 pg/L), which was set by US EPA (2003). PCDD/Fs enter the aquatic environment can

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occur with the discharge of wastewater (UNEP Chemicals, 2005), application of pesticides (Masunaga *et al.*, 2001; UNEP Chemicals, 2005) as well as dry and wet deposition from the atmosphere. But how PCDD/Fs enter tap water through deposition has seldom been investigated, a deficiency this study aims to address.

In Taiwan, there were several studies regarding the atmospheric PCDD/F deposition from relevant sources. In the study of Chi *et al.* (2009), owing to the automated PCDD/F ambient sampler can prevent both re-suspension and photo degradation of the PCDD/Fs collected, the PCDD/F deposition flux collected using the automated PCDD/F sampler was significantly higher than that sampled with the cylindrical vessels. The wet deposition flux of PCDD/Fs observed was significantly higher than the dry deposition flux, demonstrating that wet deposition is the major PCDD/F removal mechanism in the atmosphere. Shih *et al.* (2006) investigated dry deposition of PCDD/Fs in the ambient air in southern Taiwan. Atmospheric dry deposition fluxes of total PCDD/Fs averaged approximately 150 pg/m^2/day. The total dry deposition flux was found to decrease as the temperature increased. Calculated dry deposition velocities of total PCDD/Fs were averaged 0.42 cm/s. Atmospheric dry deposition of PCDD/Fs in the vicinity of municipal solid waste incinerators was investigated by Wu *et al.* (2009). Dry deposition fluxes of total PCDD/Fs were 18.0 and 23.5 pg I-TEQ/m^2-day in the ambient air near two MSWIs. They were considerably higher than those measured in Guangzhou, China. While annual dry deposition fluxes were 189 and 217 ng/m^2-year, which were also much higher than that near to the Atlantic Ocean. Notably, Asian dust storm (ADS) that originated in the deserts of Mongolia and Mainland China eventually reached Taiwan, and then significantly increased the atmospheric PCDD/F concentrations. Additionally, the amount of PCDD/Fs bound to suspended particles increased during the ADS episode (Chi *et al.*, 2008).

In this study, the data for PCDD/Fs in intake source water, tap water, and the ambient air on a water treatment plant are investigated. In addition, atmospheric dry- and wet-deposition on the surface of water treatment facilities was assessed and its influence on the level of PCDD/Fs in treated water was evaluated.

METHODS

PCDD/Fs in Source Water and Tap Water

In order to have a better understanding of PCDD/Fs in source water of DWTPs in Taiwan, a total of 17 samples were taken from a reservoir (Sin-Shan Reservoir, n = 1) located in northern Taiwan, a river located in middle Taiwan (Jhuo-Shuei River, n = 4), and a river located in southern Taiwan (Gao-Ping River, n = 12). Jhuo-Shuei River is the longest river in Taiwan, and Gao-Ping River has the largest watershed area in Taiwan.

For tap water, a total of 4 samples of treated water taken from a drinking water treatment plant (DWTP-LN) using the water of Jhuo-Shuei River as raw water. In addition, a total of 14 samples of tap water were taken from the user in a highly industrialized area using the Gao-Ping River as raw water. Because of the very low levels of PCDD/Fs present in water, an in situ pre-concentration system was used for sampling of source water (averagely 554 L, n = 17) and tap water (averagely 1360 L, n = 18). This system consisted of a pump which could be set in the range of 0.2–0.8 L/min, a glass fiber filter holder, polyurethane foam holders and a water meter to measure the volume of water sampled.

PCDD/Fs in the Ambient Air of Drinking Water Treatment Plant LN

In 2003, a total of 4 samples of ambient air were taken from the atmosphere of DWTP-LN simultaneously with the water sampling. Each ambient air sample was collected using a PS-1 sampler (Graseby Anderson, GA, USA) according to the US EPA Compendium Method TO-9A. The sampling flow rate was 0.225 m^3/min. Each sample was collected continuously on three consecutive days, yielding a sampling volume of 972 m^3. The PS-1 sampler was equipped with a quartz-fiber filter for sampling particle-phase PCDD/Fs, which was followed by a glass cartridge containing PUF for sampling gas-phase PCDD/Fs. A known amount of surrogate standard was spiked to the PUF in the laboratory before the field sampling was conducted. The recoveries of PCDD/F surrogate standards ranged between 82% and 115%. They met the criteria within 70–130%. Relative standard deviation also calculated and presented in relevant tables.

Considering that PCDD/F concentrations in the ambient air generally vary with different seasons, the sampling and analyzing of these four samples were scheduled in spring, summer, fall, and winter, respectively. Furthermore, based on the variation of meteorological conditions (such as precipitation, sunshine, temperature, etc.) near the sampling sites in the past few years, representative days were thus selected for ambient air sampling.

Analysis of PCDD/Fs

PCDD/F analyses of water and ambient air samples followed the US EPA Method-1613B and EPA Compendium Method TO-9A, respectively. All chemical analyses were conducted by the Super Micro Mass Research and Technology Center in Cheng-Shiu University, certified by the Taiwan EPA for analyzing PCDD/Fs. Each collected sample was spiked with a known amount of the internal standard solution to the extraction thimble prior to PCDD/F analysis. After being extracted for 24 h, the extract was concentrated, treated with concentrated sulphuric acid, and subjected to a series of sample cleanup and fractionation procedures. The elute was concentrated to approximately 1 mL and transferred to a vial. The concentrate was further concentrated to near dryness, using a stream of nitrogen. Immediately prior to analysis, the standard solution was added to the sample to ensure recovery during the analysis process. A high-resolution gas chromatographs/high- resolution mass spectrometers (HRGC/HGMS) were used for PCDD/F analyses. The HRGC (Hewlett Packard 6970 Series, CA, USA) was

equipped with splitless injection and a DB-5 fused silica capillary column (60 m length, 0.25 mm ID, and 0.25 μm film thickness) (J&W Scientific, CA, USA). The oven temperature program was set as follows: begin at 150°C (held for 1 min), then increase by 30°C/min to 220°C (held for 12 min), then increase by 1.5°C/min to 240°C (held for 5 min), and finally increase by 1.5°C/min to 310°C (held for 20 min). Helium was used as the carrier gas. The HRMS (Micromass Autospec Ultima, Manchester, UK) was equipped with a positive electron impact (EI+) source. The analyzer mode of the selected ion monitoring (SIM) with a resolving power at 10,000 was used. The electron energy and source temperature were specified at 35 eV and 250°C, respectively.

For analysis of PCDD/Fs, blank tests were implemented for both filter and PUF. One set of blanks, one for filter and one for PUF, were conducted for every ten actual samples. Field and laboratory blank samples were routinely analyzed for quality assurance purposes. However, the PCDD/F levels in blank tests were normally very low and not detectable in the present study. Thus blank correction was not required. For the samples collected from the ambient air, the method detection limits (MDLs) conducted in this study were 0.8 fg/m^3 for OCDD and < 0.1 fg/m^3 for other congeners; While for the samples collected from source and tap water, MDLs were 0.2 fg/L for OCDD and < 0.01 fg/L for other congeners.

Atmospheric Deposition of PCDD/Fs on the Water Surface of Treatment Facilities

Using the measured data of PCDD/Fs in the atmosphere of DWTP-LN and theoretical model, the gas/particle partition, gas/particle deposition velocity, and dry deposition flux were built up. For wet deposition, we used the gas- and particle-phase concentrations, gas scavenging ratio, and particle scavenging ratio, then the dissolved and particle phase concentrations of rain water can be obtained. Finally by taking into account the weather information gathered from the nearest weather station, the annual dry and wet deposition can be obtained.

Atmospheric deposition of PCDD/Fs on the water surface of drinking water treatment facilities will mix with the water, and a fraction of PCDD/F mass in water will be removed by a conventional treatment process. The increased concentration of PCDD/Fs in water via deposition will be compared to the level in source water and tap water.

RESULTS AND DISCUSSION

PCDD/Fs in Source Water

As shown in Table 1, the means of total PCDD/F concentrations in source water are 0.0049, 0.0086, and 0.0107 pg WHO-TEQ/L for Sin-Shan Reservoir, Jhuo-Shuei River, and Gao-Ping River, respectively. They are all much lower than the environmental water quality standard (1 pg WHO-TEQ/L) issued by the government of Japan, indicating that Taiwan government has paid much attention to improve environmental water quality. Fig. 1 shows the congener profiles of 17 PCDD/Fs in three

different source water. The mass fractions of 17 PCDD/Fs in three source water are very similar, and the top three predominant congeners are OCDD, 1,2,3,4,6,7,8-HpCDD and OCDF.

PCDD/Fs in Tap Water

Table 2 shows the mean concentrations of PCDD/Fs in tap water. The mean concentrations of total PCDD/Fs are 0.0039, 0.0013, and 0.015 pg WHO-TEQ/L for LN, GS, and RW area, respectively. They are all lower than those in the raw water before treatment and are also lower than the environmental quality standard for dioxins in drinking water (12 pg WHO-TEQ/L) proposed by Taiwan EPA (http://tsm. epa.gov.tw/drinkwater/law/law04.htm); Furthermore, they are even lower than the current maximum allowable dioxins level of 1 pg WHO-TEQ/L in Japan (Environmental Health Department, 2002). These results reveal that the purification units in the treatment plant could effectively removed PCDD/Fs. Fig. 2 shows the congener profiles of 17 PCDD/Fs in tap water. Comparing with the source water, the most predominant congener is still OCDD (Fig. 1 and 2), but the patterns in tap water (Fig. 2) are somewhat different from those in the source water (Fig. 1).

As mentioned in the METHOD part, the water source of LN area is Jhuo-Shuei River, and those of GS and RW areas are Gao-Ping River. Thus, the PCDD/F levels of tap water from LN, GS and RW area and those of the source water (Sin-Shan reservoir, Jhuo-Shuei River and Gao-Ping River) are not comparable.

PCDD/Fs in the Ambient Air of Drinking Water Treatment Plant LN

Table 3 shows the concentrations of PCDD/Fs in the ambient air of DWTP-LN. The concentrations of total PCDD/Fs ranged from 0.024 to 0.493 pg I-TEQ/Nm^3 with an average of 0.277 pg I-TEQ/Nm^3, which is approximately three times (0.277/0.088) higher than that in other area (0.088 pg I-TEQ/Nm^3) in Taiwan (Wang *et al.*, 2005). Fig. 3 shows the congener profiles of 17 PCDD/Fs for the air of DWTP-LN. The predominant congeners are OCDD, followed by 1,2,3,4,6,7,8-HpCDD, 1,2,3,4,6,7,8-HpCDF, and OCDF. The congener profile of ambient air was different from stack flue gas from a municipal solid waste incinerator or from unburned joss paper, such as OCDD, HpCDF, HpCDD, HxCDF and HxCDD (Hu *et al.*, 2009; Wang *et al.*, 2009).

Atmospheric Deposition of PCDD/Fs on the Water Surface of Treatment Facilities
Particle/gas partitions

An equation that has been used with success to describe particle-gas partitioning is as follow:

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

Where K_p (m^3/μg) is a temperature-dependent partitioning constant, TSP (μg/m^3) is the concentration of total suspended particulate material, F (pg/m^3) is the concentration of the

Table 1. PCDD/Fs in source water

Congeners	Sin-Shan Reservoir (n = 1)		Jhuo-Shuei River (n = 4)		Gao-Ping River (n = 12)	
	Ave.	RSD (%)	Ave.	RSD (%)	Ave.	RSD (%)
2,3,7,8-TeCDD	0.0000	-	0.0009	41	0.0011	55
1,2,3,7,8-PeCDD	0.0008	-	0.0014	36	0.0012	77
1,2,3,4,7,8-HxCDD	0.0000	-	0.0019	80	0.0014	79
1,2,3,6,7,8-HxCDD	0.0018	-	0.0045	83	0.0036	93
1,2,3,7,8,9-HxCDD	0.0007	-	0.0045	94	0.0031	102
1,2,3,4,6,7,8-HpCDD	0.0342	-	0.0987	98	0.0695	109
OCDD	0.146	-	1.87	76	0.7990	104
2,3,7,8-TeCDF	0.0083	-	0.0023	22	0.0059	70
1,2,3,7,8-PeCDF	0.0021	-	0.0022	31	0.0045	61
2,3,4,7,8-PeCDF	0.0032	-	0.0043	27	0.0069	91
1,2,3,4,7,8-HxCDF	0.0025	-	0.0036	43	0.0072	73
1,2,3,6,7,8-HxCDF	0.0022	-	0.0035	34	0.0052	88
1,2,3,7,8,9-HxCDF	0.0008	-	0.0007	57	0.0014	91
2,3,4,6,7,8-HxCDF	0.0029	-	0.0040	49	0.0061	85
1,2,3,4,6,7,8-HpCDF	0.0087	-	0.0276	28	0.0488	93
1,2,3,4,7,8,9-HpCDF	0.0015	-	0.0040	43	0.0040	89
OCDF	0.0162	-	0.180	78	0.243	163
PCDDs	0.184	-	1.99	77	0.879	103
PCDFs	0.0484	-	0.232	62	0.333	0.4
PCDDs/PCDFs ratio	3.80	-	8.57	-	2.64	-
Total PCDD/Fs (pg/L)	0.232	-	2.22	69	1.21	106
PCDDs (pg I-TEQ/L)	0.0011	-	0.0056	61	0.0040	71
PCDFs (pg I-TEQ/L)	0.0035	-	0.0042	26	0.0070	78
PCDDs/PCDFs (I-TEQ) ratio	0.33	-	1.33	-	0.57	-
Total TEQ (pg I-TEQ/L)	0.0046	-	0.0097	46	0.0110	75
PCDDs (pg WHO-TEQ/L)	0.0014	-	0.0046	60	0.0039	70
PCDFs (pg WHO-TEQ/L)	0.0035	-	0.0040	26	0.0068	78
PCDDs/PCDFs (WHO-TEQ) ratio	0.40	-	1.14	-	0.57	-
Total TEQ (pg WHO-TEQ/L)	0.0049	-	0.0086	46	0.0107	74

compound of interest bound to particles, and A (pg/m^3) is the gaseous concentration of the compound of interest. (Yamasaki *et al.,* 1982; Pankow, 1991; Pankow and Bidleman, 1992; Pankow, 1994). Plotting log K_p against the logarithm of the subcooled liquid vapor pressure, P_L^o, gives

$$\text{Log } K_p = m_r \times \log P_L^o + b \qquad (2)$$

Where m_r is the slope and b_r is the y-intercept of the trendline (Lohmann and Jones, 1998). Eitzer and Hites (1988) have correlated P_L^o of PCDD/Fs with gas chromatographic retention indexes (GC-RI) on a non-polar (DB-5) GC-column using p,p'-DDT as a reference standard. The correlation has been redeveloped by Hung *et al.* (2002)

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

Where P_L^o is subcooled liquid vapor pressure, RI is gas chromatographic retention indexes derived by Donnelly *et al.* (1987) and Hale *et al.* (1985), and T is ambient temperature (K). In this study, the RIs derived by Donnelly

et al. (1987) and Hale *et al.* (1985) and Eq. (3) redeveloped by Hung *et al.* (2002) were taken to generate the P_L^o values.

A complete dataset on the gas-particle partitioning of PCDD/Fs in Taiwan has been reported (Chao *et al.,* 2004). The data gave values for $m_r = -1.29$ and $b_r = -7.2$ with $R^2 = 0.94$. In this study, the trendline proposed by Chao *et al.* (2004) was taken to estimate the partitioning constant, K_p.

Table 4 lists the environmental conditions during the sampling campaign. The temperature ranged from 18.1 to 29.5°C with an average of 23.5°C, and the concentration of total suspended particulate material (TSP) ranged from 86 to 327 μg/m^3, with an average of 212 μg/m^3. In years, Asian dust storm (ADS) that originated in the deserts of Mongolia and Mainland China eventually reached populated areas of East Asia, including Taiwan. Prior to the ADS episode, the atmospheric PCDD/F concentrations were considerably lower than those measured in other Asian countries. Nevertheless, they increased 1.9–3.2 times during the ADS episode. Notably, the amount of PCDD/Fs bound to suspended particles increased from 257–259 to 339–512 pg I-TEQ/g TSP (Chi *et al.,* 2008). As shown in Table 4 of the present study, the significantly higher TSP

Table 2. PCDD/F levels in tap water

Congeners	LN area (n = 4)		GS area (n = 7)		RW area (n = 7)	
	Ave.	RSD (%)	Ave.	RSD (%)	Ave.	RSD (%)
2,3,7,8-TeCDD	0.0005	63	0.0001	23	0.0001	56
1,2,3,7,8-PeCDD	0.0006	69	0.0002	23	0.0002	37
1,2,3,4,7,8-HxCDD	0.0006	98	0.0001	35	0.0002	31
1,2,3,6,7,8-HxCDD	0.0013	58	0.0002	38	0.0004	38
1,2,3,7,8,9-HxCDD	0.0010	61	0.0002	68	0.0002	61
1,2,3,4,6,7,8-HpCDD	0.0088	60	0.0024	73	0.0022	37
OCDD	0.0416	78	0.0432	191	0.0139	68
2,3,7,8-TeCDF	0.0021	58	0.0046	74	0.0030	87
1,2,3,7,8-PeCDF	0.0023	66	0.0004	30	0.0007	61
2,3,4,7,8-PeCDF	0.0026	57	0.0005	28	0.0010	42
1,2,3,4,7,8-HxCDF	0.0031	55	0.0004	25	0.0008	51
1,2,3,6,7,8-HxCDF	0.0019	89	0.0004	28	0.0006	43
1,2,3,7,8,9-HxCDF	0.0005	94	0.0000	184	0.0001	74
2,3,4,6,7,8-HxCDF	0.0021	89	0.0004	31	0.0006	35
1,2,3,4,6,7,8-HpCDF	0.0071	59	0.0016	57	0.0026	35
1,2,3,4,7,8,9-HpCDF	0.0017	61	0.0003	39	0.0003	36
OCDF	0.0256	52	0.0090	109	0.0065	61
PCDDs	0.0543	71	0.0465	180	0.0173	61
PCDFs	0.0490	45	0.0176	63	0.0162	50
PCDDs/PCDFs ratio	1.11	-	2.64	-	1.06	-
Total PCDD/Fs (pg/L)	0.1034	52	0.0641	140	0.0335	55
PCDDs (pg I-TEQ/L)	0.0012	61	0.0003	29	0.0003	35
PCDFs (pg I-TEQ/L)	0.0025	61	0.0009	49	0.0011	51
PCDDs/PCDFs (I-TEQ) ratio	0.48	-	0.40	-	0.32	-
Total TEQ (pg I-TEQ/L)	0.0037	58	0.0012	33	0.0014	46
PCDDs (pg WHO-TEQ/L)	0.0014	60	0.0004	29	0.0004	36
PCDFs (pg WHO-TEQ/L)	0.0025	61	0.0009	50	0.0011	50
PCDDs/PCDFs (WHO-TEQ) ratio	0.59	-	0.46	-	0.42	-
Total TEQ (pg WHO-TEQ/L)	0.0039	57	0.0013	33	0.0015	47

concentrations, 327 and 230 μg/m³, were measured at LN area during fall and winter seasons. The PCDD/Fs bound to suspended particles would thus increase and then led to the relatively higher PCDD/F concentrations in the ambient air, particularly during winter (0.493 pg I-TEQ/Nm³ in Table 3). Additionally, the lower temperatures during fall and winter seasons (Table 4) tend to lower the altitude of boundary layer. Within this layer, temperature increases with altitude and result in poor diffusion of air pollutants. As a result, the higher PCDD/F concentrations were measured in the ambient air.

Based on the environmental conditions, the calculated sub-cooled liquid vapor pressure (P_L^o) and gas-particle partitioning constant (K_p) of PCDD/Fs in the ambient air of DWTP-LN during the four sampling periods are shown in Table 5. This result shows that a compound with lower chlorine numbers has a higher sub-cooled liquid vapor pressure. It also illustrates that a compound with higher chlorine number has a higher particle-gas partitioning constant. So, at constant temperature and TSP concentrations, the congeners of PCDD/Fs with a higher chlorine number will have a higher fraction of particle phase.

The percentage of particle-gas partitioning in the ambient air of DWTP-LN is shown in Table 6. The fraction of the gas phase for congener 2,3,7,8-TeCDD ranged from 0.72 to 0.97, and 0.79 to 0.98 for 2,3,7,8-TeCDF. For the congeners with the highest chlorine number, the fraction of the particle phase for congener OCDD ranged from 0.95 to 1.00 and 0.93 to 1.00 for OCDF.

Atmospheric dry deposition of PCDD/Fs

The atmospheric dry deposition flux of PCDD/Fs is a combination of both gas- and particle-phase flux, which is given by

$$F_T = F_g + F_p$$
$$C_T \times V_{d,T} = C_g \times V_{d,g} + C_p \times V_{d,p} \tag{4}$$

where F_T is the total PCDD/F deposition flux contributed by the summation of both gas- and particle-phase flux, F_g is the PCDD/F deposition flux contributed by the gas phase, F_p is the PCDD/F deposition flux contributed by the particle phase, C_T is the measured concentration of total PCDD/Fs in the ambient air, $V_{d,T}$ is the dry deposition
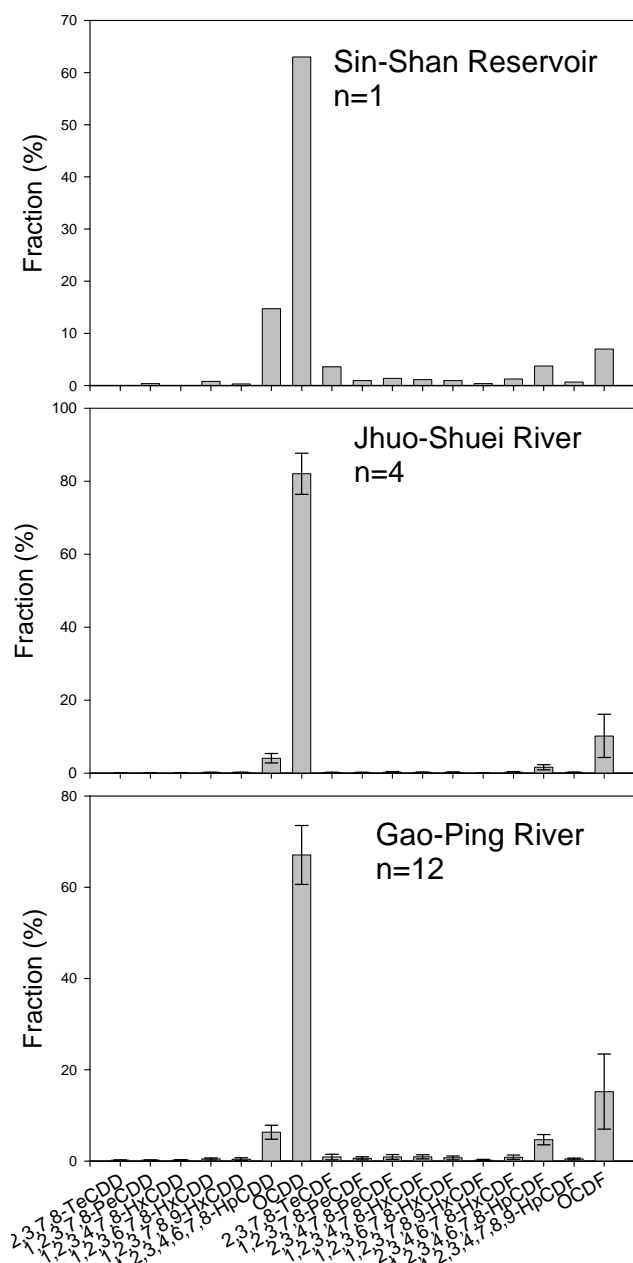
Fig. 1. Congener profiles of 17 PCDD/Fs in source water.
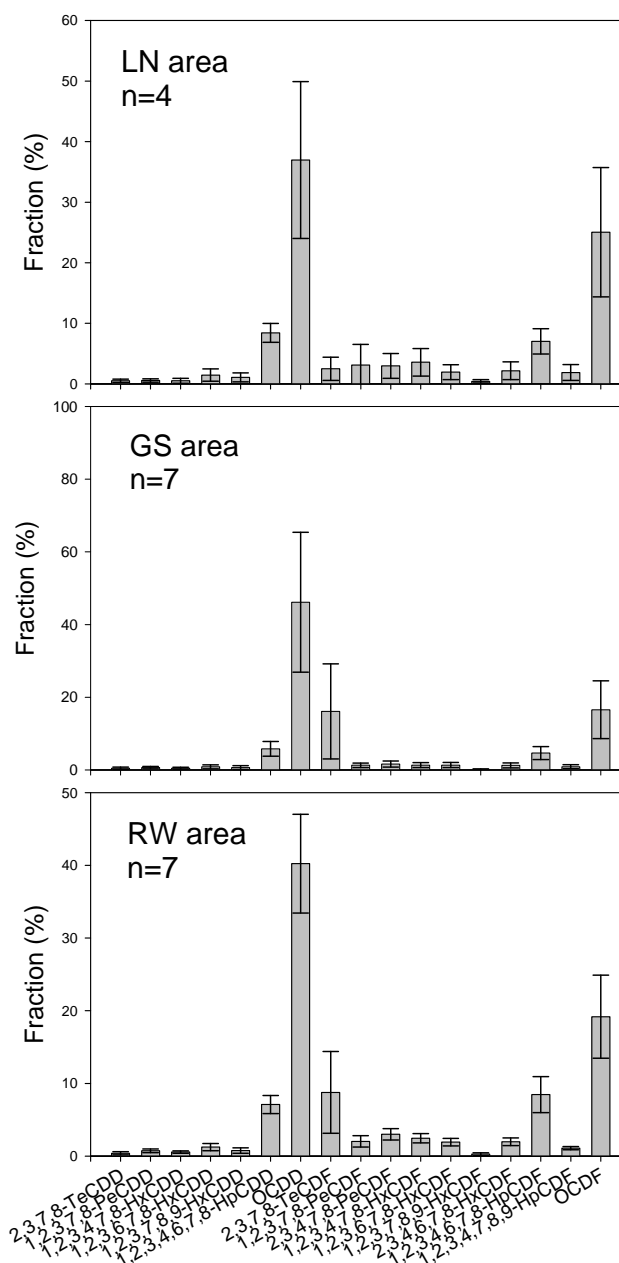
Fig. 2. Congener profiles of 17 PCDD/Fs in tap water.

velocity of total PCDD/Fs, C_g is the calculated concentration of PCDD/Fs in the gas phase, $V_{d,g}$ is the dry deposition velocity of the gas-phase PCDD/Fs, C_p is the calculated concentration of PCDD/Fs in the particle phase, and $V_{d,p}$ is the dry deposition velocity of the particle-phase PCDD/Fs.

The mean dry deposition velocity of total PCDD/Fs (0.42 cm/s) was proposed by Shih *et al.* (2006). This value ($V_{d,T}$ = 0.42 cm/s) is also used for the approximate calculation of total PCDD/F dry deposition flux.

Dry deposition of gas-phase PCDD/Fs is mainly by diffusion, because of a lack of measured data for PCDD/Fs, a selected value (0.010 cm/s) of gas-phase PAH dry deposition velocity, $V_{d,g}$, proposed by Sheu *et al.* (1996) and used by Lee *et al.* (1996) is also used here to calculate the

PCDD/F dry deposition flux contributed by its gas phase.

Dry deposition of particle-phase PCDD/Fs is mainly achieved by gravitational settling, and the dry deposition velocity of particle-phase PCDD/Fs, $V_{d,p}$, can be calculated by Eq. (4). The calculated dry deposition velocity of particle-phase PCDD/Fs, and the data bank required for this calculation, are listed in Table 7. The data shows that the mean dry deposition velocity of particle-phase PCDD/Fs is 0.44 cm/s for the ambient air of DWTP-LN.

Table 8 lists the deposition fluxes of PCDD/Fs in the ambient air of DWTP-LN. The mean dry deposition fluxes of PCDD/Fs contributed by the gas phase and particle phase are 0.769 and 71.5 pg/m²-day, respectively, and 98.9% was contributed by particle-phase deposition. These results demonstrate that the dry deposition of PCDD/Fs was

Table 3. PCDD/F concentrations in the ambient air of DWTP-LN.

Congeners	Spring	Summer	Fall	Winter	Ave.	RSD (%)
2,3,7,8-TeCDD	0.009	0.001	0.009	0.015	0.009	66
1,2,3,7,8-PeCDD	0.067	0.003	0.028	0.074	0.043	77
1,2,3,4,7,8-HxCDD	0.144	0.003	0.030	0.072	0.062	99
1,2,3,6,7,8-HxCDD	0.347	0.006	0.062	0.162	0.144	104
1,2,3,7,8,9-HxCDD	0.262	0.004	0.051	0.129	0.112	101
1,2,3,4,6,7,8-HpCDD	6.05	0.043	0.409	0.942	1.86	151
OCDD	21.8	0.150	0.824	1.45	6.06	174
2,3,7,8-TeCDF	0.064	0.015	0.085	0.162	0.082	75
1,2,3,7,8-PeCDF	0.100	0.016	0.117	0.201	0.108	70
2,3,4,7,8-PeCDF	0.169	0.022	0.170	0.426	0.197	85
1,2,3,4,7,8-HxCDF	0.262	0.019	0.198	0.442	0.230	76
1,2,3,6,7,8-HxCDF	0.214	0.018	0.189	0.415	0.209	78
1,2,3,7,8,9-HxCDF	0.013	0.001	0.012	0.025	0.013	74
2,3,4,6,7,8-HxCDF	0.227	0.019	0.204	0.515	0.241	85
1,2,3,4,6,7,8-HpCDF	1.04	0.051	0.605	1.22	0.729	71
1,2,3,4,7,8,9-HpCDF	0.147	0.012	0.113	0.228	0.125	72
OCDF	1.55	0.043	0.504	0.812	0.727	87
PCDDs	28.7	0.211	1.41	2.84	8.29	165
PCDFs	3.78	0.216	2.200	4.44	2.66	71
PCDDs/PCDFs ratio	7.6	0.98	0.64	0.64	3.1	-
Total PCDD/Fs (pg/Nm^3)	32.4	0.427	3.61	7.28	10.9	133
PCDDs	0.200	0.005	0.042	0.099	0.086	98
PCDFs	0.181	0.020	0.168	0.394	0.191	81
PCDDs/PCDFs (TEQ) ratio	1.1	0.24	0.25	0.25	0.45	-
Total TEQ (pg I-TEQ/Nm^3)	0.381	0.024	0.210	0.493	0.277	74
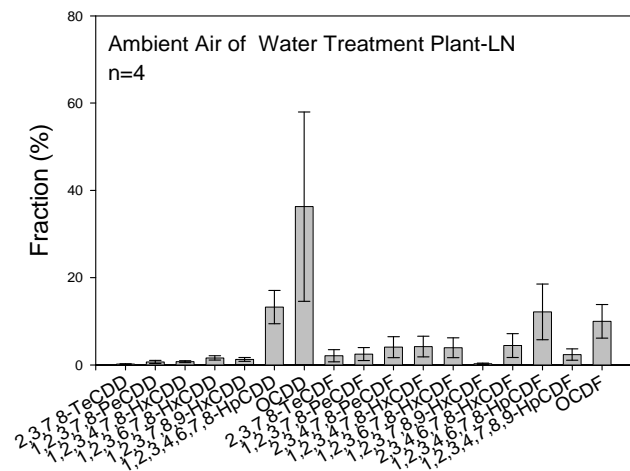
Fig. 3. Congener profiles of 17 PCDD/Fs in the ambient air of drinking water treatment plant LN.

primarily contributed by the particle phase. This is due to that the dry deposition velocity of particle-phase PCDD/Fs (0.44 cm/s) was much higher than that of gas-phase velocity (0.010 cm/s).

Atmospheric wet deposition

The wet deposition flux of PCDD/Fs was a combination of both vapor dissolution into rain and the removal of suspended particulate by precipitation. For a slightly soluble trace organic compound as PCDD/Fs, it is commonly thought that equilibrium partitioning occurs between the compound in the gas phase and a falling rain drop (Ligocki *et al.*, 1985a, b). The gas scavenging ratio, S_g, can be estimated by

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

Where S_g is the gas scavenging ratio of PCDD/Fs (dimensionless), R is the universal gas constant (82.06 × 10^{-6} m^3 atm/mol-K), T is ambient temperature (K), and H is the Henry constant (m^3 atm/mol). On the other hand, particle scavenging is not based on equilibrium considerations and depends largely on meteorological factors and particle characteristics.

Scavenging ratio is defined as the concentration of the pollutant in the raindrop divided by the concentration in the surrounding air during the precipitation event. The gas scavenging ratio, S_g, can be calculated by

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

where S_g is the gas scavenging ratio of PCDD/Fs (dimensionless), it is a ratio of the concentration of the dissolved phase in the raindrop divided by the concentrations of the gas phase in the air, $C_{rain,dis}$ is the

Table 4. Environmental conditions during the sampling campaign.

WTP	Sampling Period	Mean Temp. (°C)	TSP[a] (μg/m^3)
	3/25–3/28, 2003	21.0	204
LN	6/30–7/3, 2003	29.5	86
	11/18–11/21, 2003	25.2	327
	12/22–12/25, 2003	18.1	230

[a] TSP estimated based on average TSP concentrations gathered from an air quality monitoring station at the vicinity of WTP-LN.

Table 5. Sub-cooled vapor pressure and particle-gas partitioning constant of 17 PCDD/Fs in the ambient air of WTM-LN.

PCDD/Fs	RI[a]	Sampling periods							
		3/25–3/28, 2003		6/30–7/3, 2003		11/18–11/21, 2003		12/22–12/25, 2003	
		P_L^o (Pa)[b]	K_p (m^3/μg)[c]	P_L^o (Pa)[b]	K_p (m^3/μg)[c]	P_L^o (Pa)[b]	K_p (m^3/μg)[c]	P_L^o (Pa)[b]	K_p (m^3/μg)[c]
2,3,7,8-TeCDD	2386	5.18E-04	1.09E-03	1.40E-03	3.03E-04	8.53E-04	5.74E-04	3.65E-04	1.72E-03
1,2,3,7,8-PeCDD	2587	1.36E-04	6.11E-03	3.91E-04	1.57E-03	2.31E-04	3.10E-03	9.39E-05	9.89E-03
1,2,3,4,7,8-HxCDD	2781	3.76E-05	3.22E-02	1.14E-04	7.71E-03	6.55E-05	1.57E-02	2.54E-05	5.35E-02
1,2,3,6,7,8-HxCDD	2788	3.59E-05	3.42E-02	1.09E-04	8.16E-03	6.26E-05	1.67E-02	2.42E-05	5.69E-02
1,2,3,7,8,9-HxCDD	2802	3.27E-05	3.86E-02	9.97E-05	9.15E-03	5.72E-05	1.88E-02	2.20E-05	6.43E-02
1,2,3,4,6,7,8-HpCDD	2994	9.13E-06	2.00E-01	2.95E-05	4.41E-02	1.64E-05	9.38E-02	6.02E-06	3.42E-01
OCDD	3196	2.38E-06	1.13E+00	8.17E-06	2.31E-01	4.42E-06	5.10E-01	1.54E-06	1.98E+00
2,3,7,8-TeCDF	2338	7.13E-04	7.23E-04	1.90E-03	2.05E-04	1.16E-03	3.84E-04	5.04E-04	1.13E-03
1,2,3,7,8-PeCDF	2507	2.32E-04	3.08E-03	6.49E-04	8.17E-04	3.89E-04	1.58E-03	1.61E-04	4.93E-03
2,3,4,7,8-PeCDF	2545	1.80E-04	4.26E-03	5.10E-04	1.11E-03	3.04E-04	2.18E-03	1.25E-04	6.86E-03
1,2,3,4,7,8-HxCDF	2708	6.10E-05	1.72E-02	1.81E-04	4.24E-03	1.05E-04	8.53E-03	4.15E-05	2.84E-02
1,2,3,6,7,8-HxCDF	2714	5.86E-05	1.82E-02	1.74E-04	4.45E-03	1.01E-04	8.97E-03	3.99E-05	2.99E-02
1,2,3,7,8,9-HxCDF	2772	3.99E-05	2.98E-02	1.21E-04	7.16E-03	6.95E-05	1.46E-02	2.69E-05	4.95E-02
2,3,4,6,7,8-HxCDF	2748	4.68E-05	2.43E-02	1.41E-04	5.88E-03	8.12E-05	1.19E-02	3.17E-05	4.02E-02
1,2,3,4,6,7,8-HpCDF	2898	1.73E-05	8.79E-02	5.42E-05	2.01E-02	3.06E-05	4.19E-02	1.15E-05	1.48E-01
1,2,3,4,7,8,9-HpCDF	2986	9.62E-06	1.87E-01	3.10E-05	4.13E-02	1.73E-05	8.77E-02	6.36E-06	3.19E-01
OCDF	3147	3.30E-06	7.43E-01	1.12E-05	1.54E-01	6.08E-06	3.38E-01	2.14E-06	1.30E+00

[a] RI: Retention indexes (Donnelly *et al.*, 1987; Hale *et al.*, 1985)
[b] log P_L^o = –1.34 (RI)/T + 1.67 × 10^{-3} (RI) – 1320/T + 8.087 (Hung *et al.*, 2002)
[c] Using the equation of : log K_p = –1.29 log P_L^o – 7.2 (Chao *et al.*, 2004)

Table 6. Particle-gas partitioning in the ambient air of drinking water treatment plant LN.

PCDD/Fs	Sampling periods									
	3/25–3/28, 2003		6/30–7/3, 2003		11/18–4/21, 2003		12/22–12/25, 2003		Average	
	G[a]	P[b]	G[a]	P[b]	G[a]	P[b]	G[a]	P[b]	G[a]	P[b]
2,3,7,8-TeCDD	0.82	0.18	0.97	0.03	0.84	0.16	0.72	0.28	0.84	0.16
1,2,3,7,8-PeCDD	0.45	0.55	0.88	0.12	0.50	0.50	0.31	0.69	0.53	0.47
1,2,3,4,7,8-HxCDD	0.13	0.87	0.60	0.40	0.16	0.84	0.08	0.92	0.24	0.76
1,2,3,6,7,8-HxCDD	0.13	0.87	0.59	0.41	0.15	0.85	0.07	0.93	0.23	0.77
1,2,3,7,8,9-HxCDD	0.11	0.89	0.56	0.44	0.14	0.86	0.06	0.94	0.22	0.78
1,2,3,4,6,7,8-HpCDD	0.02	0.98	0.21	0.79	0.03	0.97	0.01	0.99	0.07	0.93
OCDD	0.00	1.00	0.05	0.95	0.01	0.99	0.00	1.00	0.02	0.98
2,3,7,8-TeCDF	0.87	0.13	0.98	0.02	0.89	0.11	0.79	0.21	0.88	0.12
1,2,3,7,8-PeCDF	0.61	0.39	0.93	0.07	0.66	0.34	0.47	0.53	0.67	0.33
2,3,4,7,8-PeCDF	0.53	0.47	0.91	0.09	0.58	0.42	0.39	0.61	0.60	0.40
1,2,3,4,7,8-HxCDF	0.22	0.78	0.73	0.27	0.26	0.74	0.13	0.87	0.34	0.66
1,2,3,6,7,8-HxCDF	0.21	0.79	0.72	0.28	0.25	0.75	0.13	0.87	0.33	0.67
1,2,3,7,8,9-HxCDF	0.14	0.86	0.62	0.38	0.17	0.83	0.08	0.92	0.25	0.75
2,3,4,6,7,8-HxCDF	0.17	0.83	0.66	0.34	0.20	0.80	0.10	0.90	0.28	0.72

Table 6. *(continued).*

PCDD/Fs	Sampling periods									
	3/25–3/28, 2003		6/30–7/3, 2003		11/18–4/21, 2003		12/22–12/25, 2003		Average	
	G[a]	P[b]	G[a]	P[b]	G[a]	P[b]	G[a]	P[b]	G[a]	P[b]
1,2,3,4,6,7,8-HpCDF	0.05	0.95	0.37	0.63	0.07	0.93	0.03	0.97	0.13	0.87
1,2,3,4,7,8,9-HpCDF	0.03	0.97	0.22	0.78	0.03	0.97	0.01	0.99	0.07	0.93
OCDF	0.01	0.99	0.07	0.93	0.01	0.99	0.00	1.00	0.02	0.98

[a] G: Gas phase; [b] P: Particle phase

dissolved-phase concentration of PCDD/Fs in the raindrop, and C_g is the concentration of PCDD/Fs in the gas phase. The particle scavenging ratio, S_p, on the other hand, can be calculated by

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

Where S_p is the particle scavenging ratio of PCDD/Fs (dimensionless), it is a ratio of the concentration of the particle phase in the raindrop divided by the concentrations of the particle phase in the air, $C_{rain,particle}$, is the particle-phase concentration of PCDD/Fs in the raindrop, and C_p is the concentration of PCDD/Fs in the particle phase.

Total scavenging of precipitation (S_{tot}) is the sum of gas and particle scavenging, it can be calculated by

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

Where S_{tot} is the total scavenging ratio of PCDD/Fs (dimensionless), and Φ is the fraction of the total air concentration bound to particles.

The particle scavenging ratio of PCDD/Fs is based on the average Bloomington air and rain concentrations that were measured by Eitzer and Hites (1989). Suppose the seventeen 2,3,7,8-substitued PCDD/Fs were evenly distributed among different particle sizes and the particles in ambient air were wash out uniformly, a common value should be acquired for all congeners. But the data measured by Eitzer and Hites (1989) exhibit a wide diversity of S_p values. There is a possibility that a congener dissolved in a raindrop is re-adsorbed by the particle scavenged. Accordingly, the S_p values of congeners which have lowest solubility for PCDD and PCDF, i.e., OCDD and OCDF, should be more accurate. Because of a lack of measured data, the S_p values of OCDD and OCDF measured by Eitzer and Hites (1989) were averaged and used here.

The scavenging ratios of PCDD/Fs in the ambient air of DWTP-LN are shown in Table 9. Among the 7 congeners of PCDDs, S_g values ranged from 5.46×10^2 (HxCDD) to 9.35×10^3 (PeCDD). For the 10 congeners of PCDFs, S_g values ranged from 1.65×10^3 (TeCDF) to 2.48×10^4 (OCDF). There is no consistent trend between S_g values and degree of chlorination. The average particle scavenging ratio of OCDD and OCDF measured by Eitzer and Hites (1989), i.e., 42,000, is used in this study.

Based on gas and particle scavenging ratios, the dissolved and particle phase concentrations of raindrops, $C_{rain,dis}$ and

$C_{rain,particle}$, can be calculated by Eqs. (6) and (7), respectively. Table 10 lists the calculated raindrop concentrations of PCDD/Fs at DWTP-LN. At WTP-LN, the mean dissolved and particle phase raindrop concentrations of PCDD/Fs are 1.65 and 438 pg/L, respectively, and the particle phase contributed 99.6% of those. The dissolved and particle phase raindrop TEQ concentrations of PCDD/Fs are 0.367 and 7.90 pg I-TEQ/L, respectively, and the particle phase contributed 95.6% of those.

Annual Dry and Wet Deposition Flux of PCDD/Fs

In order to estimate the annual deposition flux of PCDD/Fs, the information of precipitation and days of precipitation in 2003 were gathered from Jia-Yi weather station, and are listed in Table 11. The annual precipitation is 941 mm, and the annual days of precipitation are 60 days. The precipitation was not even through the year, it was concentrated in August to September. The annual deposition flux of PCDD/Fs entering the DWTP-LN is listed in Table 12. The total deposition flux of PCDD/Fs is 1439 ng/m²-year, and wet deposition contributed 10.0% of those. With regard to TEQ, the total deposition flux of PCDD/Fs is 27.0 ng I-TEQ/m²-year, and wet deposition contributed 12.0% of those. The above results reveal that dry deposition is more important than wet deposition for total deposition flux of PCDD/Fs in the ambient air of DWTP-LN. Comparison with similar semi volatile organic compounds, polycyclic aromatic hydrocarbons (PAHs), the dry deposition is the dominant disappearance mechanism during sampling period and also the high dry deposition velocities are threatening to human health (Chang *et al.*, 2003; Sahu *et al.*, 2008).

Atmospheric Deposition of PCDD/Fs on Water Surface of Treatment Facilities

The treatment capacity of DWTP-LN is 198,000 m³/day, with a water surface area of 23,840 m² to accept deposition from the atmosphere. According to the result described previously, there was a deposition flux of 27.0 ng I-TEQ/m²-year, and it will mix with the water treated. If the deposition was not removed by the treatment units, it will increase the PCDD/F concentration in water up to 8.91×10^{-3} pg I-TEQ/L, which is about 92% of PCDD/Fs in raw water. Kim *et al.* (2002) reported that the majority of PCDD/F mass are well removed (removal efficiency 87%) by conventional rapid sand filtration, thus the treated water may increase 1.16×10^{-3} pg I-TEQ/L, which is about 43% of the level in tap water. The above results indicate

Table 7. Calculated dry deposition velocity of particle-phase PCDD/Fs.

WTP	Calculated mean dry deposition flux of PCDD/Fs (pg/m²-day)			Measured mean concentrations of PCDD/Fs	Calculated mean concentrations of gas-phase PCDD/Fs	Calculated mean concentrations of particle-phase PCDD/Fs	Dry deposition velocity of total velocity of PCDD/Fs	Dry deposition velocity of gas-phase PCDD/Fs	Calculated dry deposition velocity of particle-phase PCDD/Fs
	$F_{d,T}$	$F_{d,g}$	$F_{d,p}$	C_T	C_g (pg/m³)	C_p	$V_{d,T}$ (cm/s)	$V_{d,g}$	$V_{d,p}$
LN	3955	4.5	3951	10.9	0.526	10.4	0.42	0.01	0.44

Table 8. Dry deposition flux of PCDD/Fs in the ambient air of drinking water treatment plant LN.

Congener	Season-1 $F_{d,g}$ (pg/m²-d)	Season-1 $F_{d,p}$ (pg/m²-d)	Season-1 %P	Season-2 $F_{d,g}$ (pg/m²-d)	Season-2 $F_{d,p}$ (pg/m²-d)	Season-2 %P	Season-3 $F_{d,g}$ (pg/m²-d)	Season-3 $F_{d,p}$ (pg/m²-d)	Season-3 %P	Season-4 $F_{d,g}$ (pg/m²-d)	Season-4 $F_{d,p}$ (pg/m²-d)	Season-4 %P	Average $F_{d,g}$ (pg/m²-d)	Average $F_{d,p}$ (pg/m²-d)	Average %P
2,3,7,8-TeCDD	0.066	0.646	90.7	0.010	0.011	53.4	0.065	0.536	89.2	0.092	1.59	94.6	0.058	0.697	92.3
1,2,3,7,8-PeCDD	0.258	14.1	98.2	0.024	0.142	85.6	0.121	5.39	97.8	0.194	19.5	99.0	0.149	9.78	98.5
1,2,3,4,7,8-HxCDD	0.164	47.5	99.7	0.017	0.485	96.7	0.042	9.45	99.6	0.047	25.4	99.8	0.067	20.7	99.7
1,2,3,6,7,8-HxCDD	0.376	115	99.7	0.032	0.983	96.9	0.082	19.8	99.6	0.099	57.2	99.8	0.147	48.3	99.7
1,2,3,7,8,9-HxCDD	0.255	88.4	99.7	0.021	0.737	97.2	0.062	16.7	99.6	0.071	45.9	99.8	0.102	37.9	99.7
1,2,3,4,6,7,8-HpCDD	1.25	2245	99.9	0.077	12.9	99.4	0.112	151	99.9	0.102	354	100.0	0.385	691	99.9
OCDD	0.813	8252	100.0	0.062	54.3	99.9	0.042	311	100.0	0.027	550	100.0	0.236	2292	100.0
2,3,7,8-TeCDF	0.480	3.11	86.7	0.129	0.100	43.6	0.655	3.62	84.7	1.11	12.7	92.0	0.593	4.89	89.2
1,2,3,7,8-PeCDF	0.531	14.7	96.5	0.125	0.387	75.6	0.666	15.2	95.8	0.814	40.6	98.0	0.534	17.7	97.1
2,3,4,7,8-PeCDF	0.781	29.9	97.5	0.174	0.735	80.8	0.858	26.9	96.9	1.43	99.1	98.6	0.810	39.2	98.0
1,2,3,4,7,8-HxCDF	0.501	77.6	99.4	0.122	1.95	94.1	0.451	55.4	99.2	0.508	146	99.7	0.395	70.2	99.4
1,2,3,6,7,8-HxCDF	0.393	64.1	99.4	0.111	1.87	94.4	0.415	53.6	99.2	0.455	138	99.7	0.344	64.3	99.5
1,2,3,7,8,9-HxCDF	0.016	4.34	99.6	0.008	0.216	96.4	0.018	3.71	99.5	0.017	8.70	99.8	0.015	4.24	99.7
2,3,4,6,7,8-HxCDF	0.329	71.8	99.5	0.107	2.37	95.7	0.360	61.7	99.4	0.435	177	99.8	0.308	78.1	99.6
1,2,3,4,6,7,8-HpCDF	0.475	374	99.9	0.161	12.2	98.7	0.355	214	99.8	0.300	451	99.9	0.323	263	99.9
1,2,3,4,7,8,9-HpCDF	0.032	54.5	99.9	0.023	3.53	99.4	0.033	41.5	99.9	0.026	85.5	100.0	0.029	46.3	99.9
OCDF	0.088	585	100.0	0.026	15.2	99.8	0.039	190	100.0	0.023	308	100.0	0.044	275	100.0
PCDDs	3.18	10763	100.0	0.243	69.6	99.7	0.526	514	99.9	0.632	1053	99.9	1.15	3100	100.0
PCDFs	3.63	1280	99.7	0.985	38.6	97.5	3.85	666	99.4	5.12	1465	99.7	3.39	862	99.6
PCDDs/PCDFs ratio	0.88	8.4		0.25	1.8		0.14	0.77		0.12	0.72		0.35	2.9	
PCDD/Fs	6.81	12042	99.9	1.23	108	98.9	4.38	1180	99.6	5.75	2518	99.8	4.54	3962	99.9
PCDDs (TEQ)	0.288	63.5	99.5	0.030	0.486	94.3	0.145	9.64	98.5	0.212	28.3	99.3	0.168	25.5	99.3
PCDFs (TEQ)	0.594	42.6	98.6	0.143	1.21	89.4	0.656	34.8	98.1	1.01	105	99.1	0.601	46.0	98.7
PCDDs/PCDFs (TEQ ratio)	0.48	1.5		0.21	0.40		0.22	0.28		0.21	0.27		0.28	0.61	
Total TEQ	0.882	106	99.2	0.172	1.70	90.8	0.801	44.4	98.2	1.22	134	99.1	0.769	71.5	98.9

Table 9. Washout ratio of PCDD/Fs in the ambient air of drinking water treatment plant LN.

Congeners	$S_g{}^a$	$S_p{}^b$	Season-1 $S_{tot}{}^c$	Season-1 %P^d	Season-2 $S_{tot}{}^c$	Season-2 %P^d	Season-3 $S_{tot}{}^c$	Season-3 %P^d	Season-4 $S_{tot}{}^c$	Season-4 %P^d
2,3,7,8-TeCDD	7.41E+02	4.20E+04	8.25E+03	92.7	1.79E+03	59.6	7.26E+03	91.4	1.24E+04	95.7
1,2,3,7,8-PeCDD	9.35E+03	4.20E+04	2.75E+04	84.9	1.32E+04	37.8	2.58E+04	82.0	3.20E+04	91.1
1,2,3,4,7,8-HxCDD	5.46E+02	4.20E+04	3.65E+04	99.8	1.71E+04	98.1	3.53E+04	99.7	3.89E+04	99.9
1,2,3,6,7,8-HxCDD	5.46E+02	4.20E+04	3.68E+04	99.8	1.76E+04	98.2	3.56E+04	99.8	3.91E+04	99.9
1,2,3,7,8,9-HxCDD	5.46E+02	4.20E+04	3.73E+04	99.8	1.88E+04	98.4	3.62E+04	99.8	3.94E+04	99.9
1,2,3,4,6,7,8-HpCDD	1.95E+03	4.20E+04	4.10E+04	99.9	3.36E+04	98.8	4.07E+04	99.8	4.15E+04	99.9
OCDD	3.62E+03	4.20E+04	4.18E+04	100.0	4.02E+04	99.6	4.18E+04	99.9	4.19E+04	100.0
2,3,7,8-TeCDF	1.65E+03	4.20E+04	6.84E+03	79.0	2.35E+03	30.9	6.15E+03	76.2	9.99E+03	86.9
1,2,3,7,8-PeCDF	4.90E+03	4.20E+04	1.92E+04	84.3	7.34E+03	37.6	1.76E+04	81.6	2.46E+04	90.7
2,3,4,7,8-PeCDF	4.90E+03	4.20E+04	2.22E+04	88.2	8.15E+03	45.1	2.03E+04	85.9	2.76E+04	93.1
1,2,3,4,7,8-HxCDF	1.70E+03	4.20E+04	3.31E+04	98.9	1.25E+04	90.0	3.14E+04	98.6	3.66E+04	99.4
1,2,3,6,7,8-HxCDF	1.70E+03	4.20E+04	3.34E+04	98.9	1.29E+04	90.4	3.18E+04	98.6	3.69E+04	99.4
1,2,3,7,8,9-HxCDF	1.70E+03	4.20E+04	3.63E+04	99.3	1.71E+04	93.8	3.50E+04	99.2	3.87E+04	99.6
2,3,4,6,7,8-HxCDF	1.70E+03	4.20E+04	3.52E+04	99.2	1.52E+04	92.6	3.38E+04	99.0	3.81E+04	99.6
1,2,3,4,6,7,8-HpCDF	1.74E+03	4.20E+04	3.99E+04	99.8	2.72E+04	97.7	3.93E+04	99.7	4.09E+04	99.9
1,2,3,4,7,8,9-HpCDF	1.74E+03	4.20E+04	4.10E+04	99.9	3.32E+04	98.8	4.06E+04	99.9	4.15E+04	99.9
OCDF	2.48E+04	4.20E+04	4.19E+04	99.6	4.08E+04	95.8	4.18E+04	99.5	4.19E+04	99.8

a Gas scavenging ratio, dimensionless.
b Particle scavenging ratio, dimensionless.
c The sum of gas and particle scavenging, dimensionless, $S_{tot.} = S_g(1 - \Phi) + S_p \times \Phi$.
d The percentage of particle scavenging, calculated by dividing $S_p \times \Phi$ by $S_{tot.}$

Table 10. Calculated rain concentrations of PCDD/Fs at drinking water treatment plant LN.

	Calculated rain concentration, pg/L														
Congeners	Season-1 dissolved	Season-1 particle	Season-1 %P	Season-2 dissolved	Season-2 particle	Season-2 %P	Season-3 dissolved	Season-3 particle	Season-3 %P	Season-4 dissolved	Season-4 particle	Season-4 %P	Ave dissolved	Ave particle	Ave %P
2,3,7,8-TeCDD	0.006	0.071	93	0.001	0.001	60	0.006	0.059	91	0.008	0.176	96	0.00	0.077	94
1,2,3,7,8-PeCDD	0.279	1.56	85	0.026	0.016	38	0.131	0.596	82	0.210	2.15	91	0.161	1.08	87
1,2,3,4,7,8-HxCDD	0.010	5.25	100	0.001	0.054	98	0.003	1.04	100	0.003	2.80	100	0.004	2.29	100
1,2,3,6,7,8-HxCDD	0.024	12.7	100	0.002	0.109	98	0.005	2.19	100	0.006	6.32	100	0.009	5.34	100
1,2,3,7,8,9-HxCDD	0.016	9.76	100	0.001	0.081	98	0.004	1.85	100	0.004	5.07	100	0.006	4.19	100
1,2,3,4,6,7,8-HpCDD	0.281	248	100	0.017	1.43	99	0.025	16.6	100	0.023	39.1	100	0.087	76.3	100
OCDD	0.341	912	100	0.026	6.00	100	0.018	34.4	100	0.011	60.8	100	0.099	253	100
2,3,7,8-TeCDF	0.092	0.344	79	0.025	0.011	31	0.125	0.400	76	0.212	1.41	87	0.113	0.540	83
1,2,3,7,8-PeCDF	0.301	1.62	84	0.071	0.043	38	0.378	1.68	82	0.462	4.49	91	0.303	1.96	87
2,3,4,7,8-PeCDF	0.443	3.30	88	0.099	0.081	45	0.487	2.97	86	0.810	11.0	93	0.460	4.33	90

Table 10. *(continued).*

| Congeners | Calculated rain concentration, pg/L | | | | | | | | | | | | | | |
| | Season-1 | | | Season-2 | | | Season-3 | | | Season-4 | | | Ave | | |
	dissolved	particle	% P	dissolved	particle	% P	dissolved	particle	% P	dissolved	particle	% P	dissolved	particle	% P
1,2,3,4,7,8-HxCDF	0.099	8.57	99	0.024	0.215	90	0.089	6.12	99	0.100	16.1	99	0.078	7.75	99
1,2,3,6,7,8-HxCDF	0.078	7.08	99	0.022	0.207	90	0.082	5.92	99	0.090	15.2	99	0.068	7.11	99
1,2,3,7,8,9-HxCDF	0.003	0.480	99	0.002	0.024	94	0.003	0.410	99	0.003	0.961	100	0.003	0.469	99
2,3,4,6,7,8-HxCDF	0.065	7.93	99	0.021	0.262	93	0.071	6.82	99	0.086	19.5	100	0.061	8.63	99
1,2,3,4,6,7,8-HpCDF	0.095	41.4	100	0.032	1.35	98	0.071	23.7	100	0.060	49.8	100	0.065	29.0	100
1,2,3,4,7,8,9-HpCDF	0.007	6.02	100	0.005	0.390	99	0.007	4.59	100	0.005	9.45	100	0.006	5.11	100
OCDF	0.252	64.7	100	0.075	1.68	96	0.112	21.0	99	0.067	34.0	100	0.126	30.3	100
PCDDs	0.957	1189	100	0.075	7.68	99	0.191	56.8	100	0.266	116	100	0.372	342	100
PCDFs	1.43	141	99	0.374	4.26	92	1.42	73.6	98	1.90	162	99	1.28	95.3	99
PCDDs/PCDFs ratio	0.67	8.41		0.20	1.80		0.13	0.77		0.14	0.72		0.29	2.93	
PCDD/Fs	2.391	1330	100	0.449	11.9	96	1.62	130	99	2.16	278	99	1.65	438	100
PCDDs (TEQ)	0.153	7.02	98	0.014	0.054	79	0.072	1.07	94	0.115	3.12	96	0.089	2.82	97
PCDFs (TEQ)	0.271	4.71	95	0.063	0.134	68	0.300	3.84	93	0.478	11.6	96	0.278	5.08	95
PCDDs/PCDFs (TEQ ratio)	0.56	1.49		0.23	0.40		0.24	0.28		0.24	0.27		0.32	0.61	
Total TEQ	0.425	11.7	97	0.077	0.187	71	0.373	4.90	93	0.593	14.8	96	0.367	7.90	96

Table 11. Precipitation and days of precipitation in 2003 in LN area.

Month	Precipitation (mm)	Days of Precipitation (d)
January	20.9	2
February	21.5	1
March	18.6	5
April	105.1	6
May	62.8	4
June	301.1	8
July	67.7	5
August	273.7	17
September	49.1	10
October	19.1	1
November	1.5	1
December	0	0
Total	941.1	60

that the influence of atmospheric deposition of PCDD/Fs on drinking water is of great significance and the water treatment facility with a cover is suggested.

CONCLUSIONS

The results of this study come to the following conclusions:

(1) The concentrations of PCDD/Fs in source water ranged from 0.0046 to 0.0110 pg I-TEQ/L with an average of 0.0084 pg I-TEQ/L.

(2) The concentrations of PCDD/Fs in tap water ranged from 0.0012 to 0.0037 pg I-TEQ/L with an average of 0.0021 pg I-TEQ/L, which is much smaller than that in source water.

(3) The total atmospheric deposition flux of PCDD/Fs entering the DWTP-LN was 27.0 ng I-TEQ/m^2-year, and wet and dry deposition contributed approximately 12.0% and 88%, respectively. It reveals that dry deposition is more important than wet deposition.

(4) Atmospheric deposition of PCDD/Fs will increase the level in tap water up to 8.91×10^{-3} pg I-TEQ/L, which is approximately 92% of that in source water. If a removal efficiency of 87% is achieved by conventional rapid sand filtration system, the tap water may increase 1.16×10^{-3} pg I-TEQ/L, which is about 43% of the level in tap water. These results indicate that in the DWTP-LN, the influence of atmospheric deposition of PCDD/Fs on the drinking water is of great significance, and supplying water treatment facilities with a cover is suggested.

Table 12. Deposition flux of PCDD/Fs in the ambient air of drinking water treatment plant LN.

Congener	Dry deposition (ng/m²-year)			%P	Wet deposition (ng/m²-year)			% P	Total (ng/m²-year)			%W
	Fd,g	Fd,p	Fd		dissolved	particle	Fw		Fd	Fw	F_T	
2,3,7,8-TeCDD	0.018	0.229	0.248	92.6	0.003	0.032	0.035	91.2	0.248	0.035	0.282	12.3
1,2,3,7,8-PeCDD	0.048	3.24	3.29	98.6	0.085	0.380	0.464	81.8	3.29	0.464	3.76	12.4
1,2,3,4,7,8-HxCDD	0.021	6.78	6.80	99.7	0.002	0.811	0.813	99.7	6.80	0.813	7.62	10.7
1,2,3,6,7,8-HxCDD	0.047	15.9	15.9	99.7	0.005	1.81	1.82	99.7	15.9	1.82	17.7	10.2
1,2,3,7,8,9-HxCDD	0.033	12.4	12.5	99.7	0.003	1.46	1.46	99.8	12.5	1.46	13.9	10.5
1,2,3,4,6,7,8-HpCDD	0.124	226	226	99.9	0.036	23.1	23.1	99.8	226	23.1	249	9.3
OCDD	0.076	749	749	100.0	0.040	73.1	73.1	99.9	749	73.1	822	8.9
2,3,7,8-TeCDF	0.188	1.62	1.81	89.6	0.070	0.211	0.282	75.0	1.81	0.282	2.09	13.4
1,2,3,7,8-PeCDF	0.166	5.80	5.96	97.2	0.209	0.866	1.07	80.6	5.96	1.07	7.04	15.3
2,3,4,7,8-PeCDF	0.257	13.0	13.3	98.1	0.280	1.62	1.90	85.3	13.3	1.90	15.2	12.5
1,2,3,4,7,8-HxCDF	0.123	22.9	23.1	99.5	0.054	3.35	3.40	98.4	23.1	3.40	26.5	12.8
1,2,3,6,7,8-HxCDF	0.106	21.0	21.1	99.5	0.049	3.15	3.20	98.5	21.1	3.20	24.3	13.2
1,2,3,7,8,9-HxCDF	0.005	1.38	1.38	99.7	0.002	0.220	0.223	98.9	1.38	0.223	1.60	13.9
2,3,4,6,7,8-HxCDF	0.095	25.7	25.8	99.6	0.043	3.67	3.72	98.8	25.8	3.72	29.5	12.6
1,2,3,4,6,7,8-HpCDF	0.099	85.0	85.1	99.9	0.050	13.4	13.5	99.6	85.1	13.5	98.6	13.7
1,2,3,4,7,8,9-HpCDF	0.009	14.9	14.9	99.9	0.005	2.54	2.54	99.8	14.9	2.54	17.5	14.6
OCDF	0.014	88.2	88.2	100.0	0.095	13.6	13.7	99.3	88.2	13.7	102	13.5
PCDDs	0.367	1013	1014	100.0	0.174	101	101	99.8	1014	101	1114	9.1
PCDFs	1.06	280	281	99.6	0.858	42.7	43.5	98.0	281	43.5	324	13.4
PCDDs/PCDFs ratio	0.346	3.62	3.61		0.202	2.36	2.32		3.61	2.32	5.93	
PCDD/Fs	1.43	1293	1294	99.9	1.03	143	144	99.3	1294	144	1439	10.0
PCDDs	0.053	8.37	8.42	99.4	0.047	0.934	0.981	95.2	8.42	0.981	9.40	10.4
PCDFs	0.189	15.2	15.3	98.8	0.173	2.09	2.26	92.4	15.3	2.26	17.6	12.8
PCDDs/PCDFs TEQ ratio	0.282	0.55	0.55		0.270	0.447	0.433		0.55	0.43	0.98	
Total TEQ	0.243	23.5	23.8	99.0	0.220	3.02	3.24	93.2	23.8	3.24	27.0	12.0

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