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Atmospheric $PM_{2.5}$ and Polychlorinated Dibenzo-*p*-dioxin and Dibenzofuran in a Coastal Area of Central Taiwan

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ABSTRACT

Atmospheric $PM_{2.5}$ and PCDD/Fs have received much more attention in recent years due to their harmful properties. In this study, the $PM_{2.5}$ and PCDD/Fs in the ambient air and the atmospheric dry and wet depositions were investigated in a coastal area of central Taiwan. During 2014, 2015 and 2016, the mean $PM_{2.5}$ concentrations in Yunlin were 29.6, 26.6 and 26.3 µg m^{-3}, respectively. The mean values of $PM_{2.5}/PM_{10}$ ratios were in an order of Lunbei (averaged at 0.55) > Taisi (averaged at 0.54) > Mailiao (averaged at 0.363). The modeled PCDD/Fs concentration at Yunlin were 0.0380, 0.0346 and 0.0324 pg WHO_{2005}-TEQ m^{-3} during 2014 to 2016. The observed concentration of total-PCDD/Fs-WHO_{2005}-TEQ at Yunlin in January and August were 0.089 and 0.00538 pg WHO_{2005}-TEQ m^{-3}, and the modeled concentration were 0.0598 and 0.0176 pg WHO_{2005}-TEQ m^{-3}. From 2014 to 2016, the annual average dry deposition, wet deposition and total deposition in Yunlin were 4,955, 254 and 5,209 pg WHO_{2005}-TEQ m^{-2} $year^{-1}$ in 2014; 4,524, 348 and 4,872 pg WHO_{2005}-TEQ m^{-2} $year^{-1}$ in 2015; 4,224, 518 and 4,742 pg WHO_{2005}-TEQ m^{-2} $year^{-1}$ in 2016. Among these three areas, the dry deposition fluxes were highest at Mailiao, followed by Lunbei, and the lowest at Taisi. The scavenging ratios (S_{tot}) of total-PCDD/Fs-WHO_{2005}-TEQ in Yunlin were 14,600, 13,800 and 13,300 in 2014, 2015 and 2016, respectively, and with an average of 13,900. The scavenging ratios (S_{tot}) indicated a distinctive seasonal variation, and the values in spring, summer, autumn and winter were 13,700, 6,330, 12,100 and 23,500 in Yunlin, respectively. This is due to the fact that a higher temperature will cause more fraction of PCDD/Fs in the gas phase in summer and the gas phase scavenging ratio is less than that of particle phase. The results of this work provide useful information for both further studies and environmental control strategies aimed at persistent organic compounds (POPs).

Keywords: $PM_{2.5}$; PCDD/Fs; Dry deposition; Wet deposition.

INTRODUCTION

Air pollutants, such as particulate matter (PM), heavy metals and dioxins, are mainly generated from combustion processes and adversely affect human health (Schwartz *et al.*, 1996; Huang *et al.*, 2014; Wang *et al.*, 2014; Chow *et al.*, 2015; Liu *et al.*, 2016). PM is a kind of aerosol, which is defined as a mixture of solid and aqueous species which enter the atmosphere by anthropogenic and natural pathways (Ghosh *et al.*, 2014; Chen *et al.*, 2016; Lu *et al.*, 2016; Wang *et al.*, 2017). The atmosphere is a very complex system, and it is thus very challenging to control the air quality.

Polychlorinated dibenzo-*p*-dioxin and dibenzofuran (PCDD/Fs) are considered as persistent organic pollutants (POPs), and are by-products of many thermal industrial-chemical processes (Bumb *et al.*, 1980). PCDD/Fs can remain in the environment for long periods and transport over long distances due to their physical and chemical properties. Since they were first found in the fly ash of municipal solid waste incinerators (MSWIs), researchers have paid attention to various emissions sources (Wang *et al.*, 2003; Wang *et al.*, 2010; Chi *et al.*, 2015; Cheruiyot *et al.*, 2015, 2016; Wei *et al.*, 2016; Li *et al.*, 2017). According to the number and position of the chlorines, PCDD/F emissions are detected as a mixture that includes 75 PCDDs and 135 PCDFs. The characteristics of PCCD/F congener profiles have been investigated in previous studies. There are 17 congeners with chlorine substitution in the 2,3,7,8 are much more harmful than others, and the 2,3,7,8-tetrachlorodibenzo-*p*-dioxin (TCDD) is one of the most toxic chemicals (Van den Berg *et al.*, 1998; Mitrou *et al.*, 2001).

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Due to the persistence and lipophilic properties of PCDD/Fs, the atmosphere is an important pathway for transferring PCDD/Fs via dry and wet deposition from the air to the soil and aquatic eco-systems (Welsch-Pausch *et al.*, 1995; Halsall *et al.*, 1997; Ren *et al.*, 2007), and these substances then tend to bioaccumulate in the food chain, and are particularly associated with fats (such as those in milk, eggs, meat and fish), foodstuffs of animal origin being one of the main pathways of human exposure. PCDD/Fs exposure can also occur through ingestion, inhalation and dermal contact (Shih *et al.*, 2009; Chen *et al.*, 2010). However, exposure to PCDD/Fs occur mainly through bioaccumulation the food chain (Correa *et al.*, 2006), and more than 90% of human exposure is reported to be via food (Charnley and Doull, 2005). A number of studies have measured the levels of PCDD/Fs in food samples, such as meat, milk, egg, fish and seafood, in various countries and regions (Birmingham *et al.*, 1989; McLachlan *et al.*, 1997; Hsu *et al.*, 2007; Wang and Lee, 2010; Perelló *et al.*, 2015).

The atmospheric deposition processes, including dry and wet deposition, contribute significantly to the removal of atmospheric PCDD/Fs (Koester and Hites, 1992). The dry deposition of PCDD/Fs is a combination of both gas- and particle-phase fluxes. Wet deposition is the process by which atmospheric pollutants are removed via rainfall, cloud droplets or snow (Lohmann and Jones, 1998), and this is responsible for much of the higher chlorinated homologues in the environmental sinks (Shih *et al.*, 2006; Wang *et al.*, 2010). The ambient temperature, rainfall, vapor pressure and particle size will also affect the deposition process (Wu *et al.*, 2009; Wang *et al.*, 2010; Chang *et al.*, 2004). The dry deposition fluxes of PCDD/Fs are usually higher than the wet deposition fluxes, demonstrating that dry deposition is the major PCDD/Fs removal mechanism in the atmosphere (Wang *et al.*, 2010; Tseng *et al.*, 2014; Lee *et al.*, 2016; Zhu *et al.*, 2017a, b). In Taiwan, incineration is the main method of waste disposal, and there are 22 MSWIs in operation with a daily treatment capacity of 24,000 metric tons. Many studies have examined atmospheric PCDD/Fs deposition (Wang *et al.*, 2010; Mi *et al.*, 2012; Tseng *et al.*, 2014; Chi *et al.*, 2016; Lee *et al.*, 2016) in this context, and the potential environmental impact and health risk due to the emission of PCDD/Fs are matters of public concern.

In this study, the levels of $PM_{2.5}$ and PCDD/Fs in the ambient air were measured and modeled in a coastal area of Central Taiwan. In addition, both atmospheric dry and wet depositions of PCDD/Fs were investigated and discussed.

MATERIAL AND METHOD

Sample Collection

Yunlin county is located in a coastal area of central Taiwan and has a humid subtropical climate, with an annual average temperature of 22.0°C and an annual precipitation of 1,500 mm. In this study, six sampling sites in Yunlin were chosen to detect the concentrations of 17 PCDD/Fs congeners in ambient air, and the air samples were collected using a PS-1 sampler (Graseby Andersen, GA) according to the revised U.S. EPA Reference Method TO-9A. The sampling flow rate was specified at around 0.225 m^3 min^{-1}, and each sample was collected continuously on three consecutive days. The PS-1 sampler was equipped with a quartz fiber filter for sampling particle phase PCDD/Fs, followed by a glass cartridge packed with PUF and XAD-2 for sampling the gas-phase PCDD/Fs. A known amount of surrogate standard was spiked in the glass cartridge in the laboratory before sampling. To ensure that the collected samples were free of contamination, one field blank was always taken with each sampling. In addition, three townships (Lunbei, Mailiao and Taisi) were selected to model the atmospheric total PCDD/F concentrations using the metalogical data, which was collected from a local air quality station. The locations of sample sites and the air quality information are shown in Fig. 1.

Fig. 1. Sampling sites at Lunbei, Mailiao and Taisi, the red boxes represent the three air quality station, and the circles represent the air sampling sites in Yunlin.

Analysis of PCDD/Fs

Analysis of ambient air samples for PCDD/Fs was performed according to the U.S. EPA Reference Method TO-9A. All chemical analyses were measured in the Super Micro Mass Research and Technology Centre of the Cheng Shiu University, which has passed international intercalibration standards tests for PCDD/Fs in fly ash, sediment, mother's milk, human blood, and cod liver. Each sample was spiked with a known amount of the internal standard. After being extracted for 24 hours, the extract was concentrated, treated with concentrated sulfuric acid, and then subjected to a series of sample cleanup and fractionation procedures. Sample cleanup was done using an acidic silica-gel column, an alumina column, and an activated carbon column. Consequently, the elute was concentrated to around 1 mL and then transferred to a vial, and further concentrated to near dryness with a nitrogen stream. Before analyzing PCDD/Fs, the standard solution was added into the sample to ensure recovery during the analysis process (Shih *et al.*, 2006). High-resolution gas chromatographs/high-resolution mass spectrometers (HRGC/HRMS) were used for PCDD/F analysis. The HRGC (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 (EI+) source. The analyzer mode of the selected ion monitoring was used with the resolving power set at 10,000. 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).

Gas-Particle Partitioning

Gaseous and particulate concentrations of PCDD/Fs were determined by using the gas-particle partitioning fraction multiplied by the total concentrations of PCDD/Fs. The gas-particle partitioning was simulated using an equation, proposed by several researchers, that successfully describes the gas-particle partitioning constant (Yamasaki *et al.*, 1982; Pankow, 1987; Pankow and Bidleman, 1991, 1992):

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

K_p: temperature-dependent partitioning constant (m^3 μg^{-1})
TSP: concentration of total suspended particulate matter, which was multiplied by PM_{10} concentration with 1.24 ($\mu g\ m^{-3}$)
F: concentration of the compounds of interest bound to particles ($pg\ m^{-3}$)
A: gaseous concentration of the compound of interest ($pg\ m^{-3}$).

Plotting log K_p against the logarithm of the subcooled liquid vapor pressure, P_L^o, gives

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

P_L^o: subcooled liquid vapor pressure (Pa),
m_r: cited slope,
b_r: cited y-intercept.

Complete datasets on the gas-particle partitioning of PCDD/Fs in Taiwan have been reported (Chao *et al.* 2004), with the values $m_r = -1.29$ and $b_r = -7.2$ with $R^2 = 0.94$. These values were used in this study for establishing the partitioning constant (K_p) of PCDD/Fs.

A previous study correlated the 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 as follows (Hung *et al.*, 2002).

$$\log P_L^o = \frac{-1.34\,(RI)}{T} + 1.67 \times 10^{-3}(RI) - \frac{1320}{T} + 8.087 \quad (3)$$

RI: gas chromatographic retention indexes developed by Donnelly *et al.* (1987) and Hale *et al.* (1985),
T: ambient temperature (K).

Atmospheric Dry Deposition of PCDD/Fs

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

$$F_{d,T} = F_{d,g} + F_{d,p}$$
$$C_T \times V_{d,T} = C_g \times V_{d,g} + C_p \times V_{d,p} \quad (4)$$

$F_{d,T}$: the total PCDD/F deposition flux contributed by the summation of both gas- and particle-phase fluxes;
$F_{d,g}$: the PCDD/F deposition flux contributed by the gas phase;
$F_{d,p}$: the PCDD/F deposition flux contributed by the particle phase;
C_T: the measured concentration of total PCDD/Fs in the ambient air;
$V_{d,T}$: the dry deposition velocity of total PCDD/Fs;
C_g: the calculated concentration of PCDD/Fs in the gas phase;
$V_{d,g}$: the dry deposition velocity of the gas-phase PCDD/Fs;
C_p: the calculated concentration of PCDD/Fs in the particle phase;
$V_{d,p}$: the dry deposition velocity of the particle-phase PCDD/Fs.

In this study, the mean dry deposition velocity of total PCDD/Fs ($V_{d,T}$ = 0.42 cm s^{-1}) was proposed by Shih *et al.* (2006). The dry deposition of gas-phase PCDD/Fs is mainly by diffusion. Due to a lack of measured data for PCDD/Fs, a selected value (0.010 cm s^{-1}) 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).

Scavenging Ratio

For slightly soluble trace organic compounds, such 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 scavenging ratio is defined as the concentration of the pollutant in the raindrop divided by the concentration in the surrounding air during precipitation. The gas scavenging ratio, S_g, can be estimated by:

$$S_g = RT/H \tag{5}$$

S_g: the gas scavenging ratio of PCDD/Fs (dimensionless);
R: the universal gas constant (82.06×10^{-6} m^3 atm mol^{-1} K^{-1});
T: ambient temperature (K);
H: the Henry constant (m^3 atm mol^{-1}).

On the other hand, particle scavenging largely depends on meteorological factors and particle characteristics. The gas scavenging ratio is a ratio of the concentration of the dissolved phase in the raindrop divided by the concentrations of the gas phase in the air, S_g, and can be calculated by:

$$S_g = C_{rain,dis}/C_g \tag{6}$$

S_g: the gas scavenging ratio of PCDD/Fs (dimensionless);
$C_{rain,dis}$: the dissolved-phase concentration of PCDD/Fs in the raindrop;
C_g: the concentration of PCDD/Fs in the gas phase.

The particle scavenging ratio is a ratio of the concentration of the particle phase in the raindrop divided by the concentrations of the particle phase in the air, S_p, and can be calculated by:

$$S_p = C_{rain,particle}/C_p \tag{7}$$

where S_p: the particle scavenging ratio of PCDD/Fs (dimensionless);
$C_{rain,particle}$: the particle-phase concentration of PCDD/Fs in the raindrop;
C_p: the concentration of PCDD/Fs in the particle phase.

Total scavenging via precipitation is the sum of gas and particle scavenging, S_{tot}, and can be calculated by:

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

S_{tot}: the total scavenging ratio of PCDD/Fs (dimensionless);
Φ: the fraction of the total concentration bound to particles.

Because of a lack of measured data for the particle scavenging ratios of PCDD/Fs, the S_p (S_p was 42,000) values of OCDD and OCDF measured by Eitzer and Hites (1989) were averaged and used here.

Wet Deposition

Wet deposition is the removal of particles in the atmosphere by precipitation (rainfall and cloud droplets), and precipitation scavenging accounts for the majority of removing PCDD/Fs from the atmosphere by wet deposition (Huang, 2011). The wet deposition flux of PCDD/Fs is a combination of both vapor dissolution into rain and removal of suspended particulates by precipitation (Bidleman, 1988; Koester and Hites, 1992).

The wet deposition fluxes of PCDD/Fs can be evaluated by:

$$F_{w,T} = F_{w,dis} + F_{w,p} \tag{9}$$

$$F_{w,dis} = C_{rain,dis} \times Rainfall \tag{10}$$

$$F_{w,p} = C_{rain,particle} \times Rainfall \tag{11}$$

$F_{w,T}$: the wet deposition flux of PCDD/Fs from both vapor dissolution into rain and removal of suspended particulates by precipitation;
$F_{w,dis}$: the wet deposition flux contributed by vapor dissolution into rain;
$F_{w,p}$: the wet deposition flux contributed by removal of suspended particulates by precipitation;
Rainfall: monthly rainfall (m).

RESULTS AND DISCUSSION

$PM_{2.5}$ Concentration and $PM_{2.5}/PM_{10}$ Ratio

The concentration of $PM_{2.5}$ in ambient air can influence the concentration and gas-particle partition of PCDD/Fs. The meteorological data in various areas of Yunlin, Taiwan, during the period between 2014 and 2016 is presented in Tables 1–3. Among the three areas, the highest $PM_{2.5}$ concentration was found in Lunbei Township during 2014 to 2016, while other two areas had similar $PM_{2.5}$ concentrations. At Lunbei, the highest $PM_{2.5}$ concentration occurred in January (51.8 and 47.1 µg m^{-3} in 2014 and 2015, respectively) and March (41.8 µg m^{-3} in 2016), and the lowest occurred in June (15.1, 15.8 and 15.5 µg m^{-3} during 2014 to 2016). The mean concentrations of $PM_{2.5}$ were 31.4, 31.1 and 29.3 µg m^{-3} in 2014, 2015 and 2016, respectively. Mailiao and Taisi townships have the same trend as Lunbei during 2014 and 2016, as the highest $PM_{2.5}$ concentrations occurred in January in 2014 and 2015 and March in 2016, the lowest occurred in June from 2014 to 2016. In Mailiao, the mean $PM_{2.5}$ concentrations were 27.7, 24.4 and 24.9 µg m^{-3} in 2014, 2015 and 2016, respectively. While at Taisi, the mean $PM_{2.5}$ concentrations were 29.6, 24.4 and 24.7 µg m^{-3} in 2014, 2015 and 2016, respectively. From 2014 to 2016, the mean $PM_{2.5}$ concentrations in Lunbei, Mailiao and Taisi were 30.6, 25.7 and 26.2 µg m^{-3}. Overall, the mean $PM_{2.5}$ concentrations in Yunlin are 29.6, 26.6 and 26.3 µg m^{-3} in 2014, 2015 and 2016, respectively.

The $PM_{2.5}/PM_{10}$ ratio reflects the proportion of particle matter in the ambient air. The ratio $PM_{2.5}/PM_{10}$ varies widely between different areas from 2014 to 2016. As the results show, the $PM_{2.5}/PM_{10}$ ratio at Lunbei ranged between 0.383 and 0.592, 0.509 and 0.65, 0.491 and 0.663, with an average of 0.499, 0.577 and 0.582 in 2014, 2015 and 2016, respectively. For Mailiao, the $PM_{2.5}/PM_{10}$ were in a range of 0.252–0.537, 0.145–0.432, 0.261–0.498 and with an average of 0.373, 0.337 and 0.38 in 2014, 2015 and 2016, respectively. At Taisi, the $PM_{2.5}/PM_{10}$ were in the range of 0.41–0.699, 0.366–0.58, 0.389–0.622 and with an average of 0.548, 0.515 and 0.546 in 2014, 2015 and 2016,

Table 1. Meteorological data at Lunbei.

Year unit	Month	Temperature °C	PM_{10} μg m^{-3}	$PM_{2.5}$ μg m^{-3}	$PM_{2.5}/PM_{10}$	TSP μg m^{-3}	Wind speed m s^{-1}	Rainfall mm	Rainy days
2014	Jan.	17.1	102.3	51.8	0.51	126.9	3.23	0.4	2
	Feb.	17.8	62.8	36.7	0.58	77.9	3.24	17.4	11
	Mar.	19.8	77.5	45.5	0.59	96.1	2.79	49.0	7
	Apr.	23.6	66.0	39.1	0.59	81.9	2.49	11.2	5
	May	26.0	37.9	21.9	0.58	47.0	2.38	423.4	20
	June	28.3	39.3	15.1	0.38	48.7	2.97	217.6	16
	July	29.8	41.0	19.1	0.47	50.8	2.92	99.8	6
	Aug.	28.7	28.1	12.8	0.46	34.9	2.55	217.2	11
	Sep.	28.9	45.8	21.9	0.48	56.8	2.00	17.4	6
	Oct.	24.8	85.1	37.0	0.43	105.6	2.84	0.0	0
	Nov.	22.6	80.5	37.2	0.46	99.9	2.67	1.0	2
	Dec.	17.1	88.2	39.2	0.45	109.4	3.41	17.6	9
Annual		**23.7**	**62.9**	**31.4**	**0.50**	**78.0**	**2.79**	**1072.0**	**95**
2015	Jan.	16.8	79.1	47.1	0.60	98.1	3.08	9.2	5
	Feb.	17.7	76.9	44.4	0.58	95.4	2.93	17.6	3
	Mar.	20.4	64.2	37.8	0.59	79.6	2.76	5.8	5
	Apr.	23.9	53.5	28.0	0.52	66.4	2.88	43.6	6
	May	26.0	36.6	22.3	0.61	45.4	2.41	397.8	13
	June	28.8	27.3	15.8	0.58	33.8	2.55	24.4	5
	July	28.5	37.1	20.7	0.56	46.0	2.90	66.0	11
	Aug.	26.8	26.8	16.2	0.60	33.3	2.98	366.0	14
	Sep.	26.5	48.1	31.2	0.65	59.6	2.73	192.0	4
	Oct.	27.5	57.5	36.1	0.63	71.3	2.51	27.8	4
	Nov.	25.8	74.3	40.5	0.54	92.2	2.65	1.2	1
	Dec.	20.2	64.8	33.0	0.51	80.4	3.46	29.4	4
Annual		**24.1**	**53.9**	**31.1**	**0.58**	**66.8**	**2.82**	**1180.8**	**75**
2016	Jan.	16.9	49.1	30.8	0.63	60.9	3.67	143.0	15
	Feb.	15.6	63.9	35.7	0.56	79.2	3.83	29.8	7
	Mar.	17.6	63.0	41.8	0.66	78.2	2.89	127.6	15
	Apr.	25.1	59.2	36.7	0.62	73.4	2.43	115.6	9
	May	27.6	41.6	25.5	0.61	51.6	2.53	40.6	9
	June	29.1	30.8	15.5	0.50	38.2	2.68	208.2	16
	July	29.5	31.8	18.9	0.60	39.4	2.85	115.2	13
	Aug.	29.0	37.1	23.6	0.64	46.1	2.08	133.4	12
	Sep.	27.6	37.6	19.6	0.52	46.6	3.06	85.4	12
	Oct.	27.0	56.8	33.8	0.60	70.4	2.42	13.6	7
	Nov.	22.8	62.9	35.6	0.57	78.0	2.84	51.8	6
	Dec.	18.4	70.5	34.6	0.49	87.4	3.33	8.2	1
Annual		**23.9**	**50.4**	**29.3**	**0.58**	**62.4**	**2.89**	**1072.4**	**122**

respectively. The mean values of the $PM_{2.5}/PM_{10}$ ratio are in the order of Lunbei (averaged at 0.553) > Taisi (averaged at 0.536) > Mailiao (averaged at 0.363).

Simulated and Measured PCDD/F Concentrations in the Ambient Air

Previous studies demonstrate that there is a strong correlation between PM_{10} level and the PCDD/Fs mass concentration (Wang *et al.*, 2010; Lee *et al.*, 2016). Based on the PM_{10} and the regression analysis, the concentrations of total-PCDD/Fs-WHO_{2005}-TEQ are modeled and presented in Table 4.

In 2014, the monthly average concentrations of total-PCDD/Fs-WHO_{2005}-TEQ were in the range of 0.0173–0.0595, 0.0209–0.0628, 0.0146–0.0605 pg WHO_{2005}-TEQ m^{-3}, with an average of 0.0376, 0.0441 and 0.0324 pg WHO_{2005}-TEQ m^{-3} at Lunbei, Mailiao and Taisi, respectively. The monthly average concentrations of total-PCDD/Fs-WHO_{2005}-TEQ in 2015 were lower than in 2014, and in the range of 0.0165–0.0462, 0.0252–0.0613, 0.016–0.0412 pg WHO_{2005}-TEQ m^{-3} and with an average of 0.0323, 0.0432 and 0.0284 pg WHO_{2005}-TEQ m^{-3} at Lunbei, Mailiao and Taisi, respectively. While in 2016, the monthly average concentration of total-PCDD/Fs-WHO_{2005}-TEQ were lower than those in 2015, in a range of 0.0189–0.0412, 0.0254–0.0574, 0.0175–0.0381 pg WHO_{2005}-TEQ m^{-3}, with an average of 0.0304, 0.0393 and 0.0274 pg WHO_{2005}-TEQ m^{-3} at Lunbei, Mailiao and Taisi, respectively. Combining the data for these three townships, the mean PCDD/F concentrations at Yunlin is 0.0380, 0.0346 and 0.0324 pg WHO_{2005}-TEQ m^{-3}

Table 2. Meteorological data at Mailiao.

Year unit	Month	Temperature °C	PM_{10} µg m^{-3}	$PM_{2.5}$ µg m^{-3}	$PM_{2.5}/PM_{10}$	TSP µg m^{-3}	Wind speed m s^{-1}	Rainfall mm	Rainy days
2014	Jan.	16.1	101.9	43.9	0.43	126.3	4.11	0.0	0
	Feb.	16.5	69.1	32.9	0.48	85.6	3.94	7.0	10
	Mar.	19.7	81.5	40.4	0.50	101.1	3.38	46.4	6
	Apr.	24.1	73.2	39.3	0.54	90.8	3.01	9.0	4
	May	25.9	43.9	18.8	0.43	54.5	2.49	309.0	18
	June	27.8	38.2	9.6	0.25	47.3	3.06	253.0	14
	July	29.1	40.4	12.1	0.30	50.1	2.85	98.6	7
	Aug.	28.8	34.2	11.0	0.32	42.4	2.45	192.0	13
	Sep.	29.5	65.5	18.5	0.28	81.2	2.21	6.8	4
	Oct.	25.6	127.1	37.1	0.29	157.6	3.82	0.0	0
	Nov.	23.3	109.2	34.6	0.32	135.4	3.48	0.4	1
	Dec.	18.7	108.1	34.3	0.32	134.0	4.50	8.4	7
Annual		**23.8**	**74.3**	**27.7**	**0.37**	**92.2**	**3.27**	**930.6**	**84**
2015	Jan.	17.3	105.4	41.2	0.39	130.7	4.14	14.2	6
	Feb.	17.9	97.4	37.8	0.39	120.7	3.72	17.4	2
	Mar.	20.1	84.7	36.6	0.43	105.0	3.46	6.6	7
	Apr.	22.8	71.9	27.8	0.39	89.2	3.35	58.8	4
	May	25.7	53.5	20.2	0.38	66.4	2.51	412.8	14
	June	28.6	41.4	6.0	0.15	51.4	2.78	39.0	5
	July	28.0	50.4	11.3	0.22	62.6	3.01	129.0	12
	Aug.	27.4	45.1	8.8	0.19	55.9	2.83	355.4	15
	Sep.	26.9	65.9	18.7	0.28	81.7	3.04	204.0	4
	Oct.	26.7	81.0	25.8	0.32	100.4	3.11	22.6	2
	Nov.	25.3	86.3	31.1	0.36	107.0	3.24	1.6	1
	Dec.	20.8	85.6	27.3	0.32	106.1	4.49	27.4	6
Annual		**24.0**	**72.4**	**24.4**	**0.34**	**89.8**	**3.31**	**1288.8**	**78**
2016	Jan.	17.8	64.5	28.1	0.44	80.0	4.55	155.0	17
	Feb.	16.9	76.3	30.0	0.39	94.6	4.82	29.4	6
	Mar.	18.4	75.3	37.5	0.50	93.4	3.36	135.2	15
	Apr.	24.2	68.0	33.0	0.49	84.4	2.57	104.2	10
	May	27.0	55.5	22.0	0.40	68.8	2.72	54.2	7
	June	28.5	41.7	10.9	0.26	51.7	2.73	186.0	12
	July	29.3	43.8	13.0	0.30	54.4	2.93	241.6	13
	Aug.	29.9	54.3	19.4	0.36	67.4	2.08	138.8	12
	Sep.	28.6	52.6	15.9	0.30	65.2	3.22	212.2	16
	Oct.	28.2	74.1	25.9	0.35	91.9	2.71	15.8	4
	Nov.	23.9	81.9	31.1	0.38	101.5	3.37	47.8	7
	Dec.	21.2	98.6	32.1	0.33	122.3	4.26	7.8	1
Annual		**24.5**	**65.6**	**24.9**	**0.38**	**81.3**	**3.28**	**1328.0**	**120**

from 2014 to 2016. These results indicate that even if it was a slow process, the concentration of total-PCDD/Fs-WHO_{2005}-TEQ decreased year by year from 2014 to 2016.

During 2014, six sampling sites in Yunlin were chosen to measure the concentrations of 17 PCDD/F congeners, and the results are shown in Table 5. In 2014, the mass concentrations of total PCDD/Fs in January were higher than those in August at all sample sites. In January 2014, the PCDD/Fs concentrations at QT, ML1, ML2, TX1, TX2 and TX3 were 1.32, 3.03, 5.26, 1.46, 1.3 and 1.32 pg m^{-3}, respectively; while in August, the PCDD/Fs concentrations were 1.32, 3.03, 5.26, 1.46, 1.3 and 1.32 pg m^{-3}. Overall, the PCDD/Fs concentrations in Yunlin in January and August were 2.28 and 0.181 pg m^{-3}. Fig. 2 show the mass fractions of 17 PCDD/Fs in the six areas. In January, the PCDD/F congener profiles of the ambient air samples are largely dominated by higher chlorinated PCDF congeners, such as OCDF and 1,2,3,4,6,7,8-HpCDF, then higher chlorinated PCDD congeners, such as OCDD and 1,2,3,4,6,7,8-HpCDD. By August, higher chlorinated PCDD/F congeners like OCDD and OCDF are predominant in the ambient air. The concentrations of PCDDs are higher than those of PCDFs in both January and August. As the results shown, the TEQ characteristic of 17 PCDD/F congeners are similar in January and August 2014. The PCDD/F congener profiles of the ambient air samples are largely dominated by 2,3,4,7,8-PeCDF. Those results are different from the mass, it may be due to the different WHO_{2005}-TEF value, which the higher chlorinated PCDF and PCDD congeners like PCDD and PCDF have a lower WHO_{2005}-TEF value (0.0003).

Table 3. Meteorological data at Taisi.

Year unit	Month	Temperature °C	PM_{10} µg m^{-3}	$PM_{2.5}$ µg m^{-3}	$PM_{2.5}/PM_{10}$	TSP µg m^{-3}	Wind speed m s^{-1}	Rainfall mm	Rainy days
2014	Jan.	17.2	104.0	42.7	0.41	128.9	6.39	0.6	3
	Feb.	17.7	58.0	31.2	0.54	72.0	6.15	18.2	11
	Mar.	20.1	70.1	41.8	0.60	86.9	5.18	66.6	7
	Apr.	24.0	53.3	36.5	0.68	66.0	4.79	10.4	3
	May	26.6	34.6	24.2	0.70	42.8	3.41	403.6	18
	June	28.5	26.3	13.8	0.52	32.6	4.15	402.8	13
	July	30.1	28.3	19.0	0.67	35.1	4.13	209.2	8
	Aug.	29.2	23.5	15.9	0.68	29.2	3.23	184.0	13
	Sep.	29.6	34.5	23.6	0.68	42.8	3.54	10.0	5
	Oct.	25.3	75.1	38.4	0.51	93.2	6.14	0.6	2
	Nov.	23.4	65.5	33.2	0.51	81.2	5.80	2.4	8
	Dec.	18.0	75.0	35.2	0.47	93.0	7.40	33.6	12
Annual		**24.2**	**54.0**	**29.6**	**0.55**	**67.0**	**5.03**	**1342.0**	**103**
2015	Jan.	17.0	70.5	39.2	0.56	87.4	6.80	20.4	8
	Feb.	18.0	67.1	35.7	0.53	83.2	6.08	18.0	5
	Mar.	20.8	54.2	30.2	0.56	67.2	5.45	10.6	6
	Apr.	24.2	44.1	22.8	0.52	54.7	5.13	78.0	4
	May	26.9	33.6	18.3	0.54	41.7	3.48	434.4	13
	June	28.7	26.3	9.6	0.37	32.6	3.37	46.2	7
	July	28.0	33.4	15.1	0.45	41.5	4.22	216.6	12
	Aug.	27.3	26.0	10.3	0.40	32.2	4.06	589.8	16
	Sep.	26.7	41.3	24.0	0.58	51.2	4.65	240.0	5
	Oct.	25.7	56.5	28.6	0.51	70.0	5.18	14.4	4
	Nov.	23.7	59.4	32.5	0.55	73.6	5.22	1.6	2
	Dec.	19.2	56.2	26.4	0.47	69.6	6.90	42.2	4
Annual		**23.8**	**47.4**	**24.4**	**0.52**	**58.7**	**5.04**	**1712.2**	**86**
2016	Jan.	16.7	46.5	27.9	0.60	57.7	7.03	211.4	17
	Feb.	16.3	53.6	27.3	0.51	66.5	7.40	50.6	8
	Mar.	18.5	58.2	35.9	0.62	72.2	5.23	205.0	17
	Apr.	24.7	51.8	32.2	0.62	64.2	3.55	144.8	10
	May	27.3	42.7	22.3	0.52	52.9	3.73	46.4	5
	June	28.9	28.6	11.1	0.39	35.4	3.45	246.2	13
	July	29.3	30.3	14.0	0.46	37.5	3.77	418.2	13
	Aug.	29.1	35.4	20.5	0.58	44.0	3.21	124.4	11
	Sep.	27.6	30.7	17.5	0.57	38.1	4.58	298.4	16
	Oct.	27.6	47.0	25.4	0.54	58.3	4.23	29.8	5
	Nov.	23.6	57.3	30.8	0.54	71.1	5.46	74.0	7
	Dec.	20.2	61.0	31.7	0.52	75.6	6.85	13.0	2
Annual		**24.2**	**45.3**	**24.7**	**0.55**	**56.1**	**4.87**	**1862.2**	**124**

Comparing the concentrations of total-PCDD/Fs-WHO_{2005}-TEQ between simulated and measured values, the observed values at Mailiao (0.114 and 0.158 pg WHO_{2005}-TEQ m^{-3} at ML1 and ML2, respectively) are approximately twice the simulated value (0.0593 pg WHO_{2005}-TEQ m^{-3}) in January, but only one fourth (the measured value in ML1 and ML2 is 0.0052 and 0.00547 pg WHO_{2005}-TEQ m^{-3}) of the simulated value (0.0181 pg WHO_{2005}-TEQ m^{-3}) in August. At Taisi, the observed values (0.0629, 0.0743 and 0.049 pg WHO_{2005}-TEQ m^{-3} in TX1, TX2 and TX3, respectively) and simulated values (0.0585 pg WHO_{2005}-TEQ m^{-3}) are similar in January (the observed values are 0.063, 0.074 and 0.049 pg WHO_{2005}-TEQ m^{-3} at TX1, TX2 and TX3, respectively), and four times (the observed values in TX1, TX2 and TX3 are 0.00527, 0.00594 and 0.00505 pg WHO_{2005}-TEQ m^{-3}) higher than the simulated values (0.0146 pg WHO_{2005}-TEQ m^{-3}) in August. The observed concentrations of total-PCDD/Fs-WHO_{2005}-TEQ at Yunlin in January and August are 0.089 and 0.00538 pg WHO_{2005}-TEQ m^{-3}, and the modeled concentrations are 0.0598 and 0.0176 pg WHO_{2005}-TEQ m^{-3}.

Dry and Wet Deposition

The dry and wet depositions were modeled using the meteorological data and the concentrations of PCDD/Fs in the atmosphere. As shown in Figs. 3 to. 5, the monthly dry deposition fluxes of total-PCDD/Fs-WHO_{2005}-TEQ decreased year by year from 2014 to 2016 at the three areas. At Lunbei, the annual dry deposition fluxes of total-PCDD/Fs-WHO_{2005}-TEQ were 4880, 4210 and 3970 pg WHO_{2005}-TEQ m^{-2} $year^{-1}$ in 2014, 2015 and 2016, respectively. While

Table 4. Modeled atmospheric total-PCDD/F-WHO_{2005}-TEQ concentration in various townships (2014–2016).

Year	Month	Lunbei	Mailiao	Taisi
2014	Jan.	0.0576	0.0573	0.0585
	Feb.	0.0356	0.0391	0.0329
	Mar.	0.0462	0.0486	0.0418
	Apr.	0.0394	0.0436	0.0319
	May	0.0229	0.0264	0.0209
	June	0.0208	0.0202	0.0141
	July	0.0216	0.0214	0.0151
	Aug.	0.0150	0.0181	0.0126
	Sep.	0.0285	0.0404	0.0216
	Oct.	0.0523	0.0778	0.0463
	Nov.	0.0495	0.0669	0.0404
	Dec.	0.0497	0.0608	0.0424
	Average	**0.0366**	**0.0434**	**0.0315**
	Range	**0.0150–0.0576**	**0.0181–0.0669**	**0.0126–0.0585**
2015	Jan.	0.0446	0.0593	0.0399
	Feb.	0.0434	0.0548	0.0379
	Mar.	0.0384	0.0504	0.0325
	Apr.	0.0321	0.0429	0.0265
	May	0.0221	0.0321	0.0204
	June	0.0146	0.0219	0.0141
	July	0.0196	0.0265	0.0177
	Aug.	0.0143	0.0238	0.0139
	Sep.	0.0298	0.0406	0.0257
	Oct.	0.0356	0.0498	0.0349
	Nov.	0.0458	0.0531	0.0367
	Dec.	0.0367	0.0482	0.0319
	Average	**0.0314**	**0.0420**	**0.0277**
	Range	**0.0143–0.0458**	**0.0219–0.0593**	**0.0139–0.0399**
2016	Jan.	0.0279	0.0365	0.0265
	Feb.	0.0362	0.0431	0.0304
	Mar.	0.0377	0.0449	0.0348
	Apr.	0.0354	0.0406	0.0311
	May	0.0251	0.0332	0.0257
	June	0.0164	0.0220	0.0152
	July	0.0169	0.0231	0.0161
	Aug.	0.0197	0.0285	0.0188
	Sep.	0.0235	0.0326	0.0193
	Oct.	0.0351	0.0457	0.0292
	Nov.	0.0388	0.0504	0.0354
	Dec.	0.0399	0.0555	0.0345
	Average	**0.0294**	**0.0380**	**0.0264**
	Range	**0.0164–0.0399**	**0.022–0.0555**	**0.0152–0.0354**

the annual temperatures were similar from 2014 to 2016 (23.7, 24.1 and 23.9°C in 2014, 2015 and 2016), the decrease in dry deposition may due to the reduction in pollution emissions from the related sources. The annual wet depositions are 263, 269 and 397 pg WHO_{2005}-TEQ m^{-2} $year^{-1}$ in 2014, 2015 and 2016. In total, the annual total depositions (dry deposition + wet deposition) were 5,140, 4,480 and 4,360 pg WHO_{2005}-TEQ m^{-2} $year^{-1}$ in 2014, 2015 and 2016, respectively. During 2014–2016, the annual average dry deposition, wet deposition and total deposition were 4,350, 310 and 4,660 pg WHO_{2005}-TEQ m^{-2} $year^{-1}$, and the contribution of dry deposition to total deposition was approximately 93.4%.

At Mailiao, the annual dry deposition fluxes of total-PCDD/Fs-WHO_{2005}-TEQ were 5,760, 5,640 and 5,130 pg WHO_{2005}-TEQ m^{-2} $year^{-1}$ in 2014, 2015 and 2016, respectively. The annual wet depositions were 248, 461 and 608 pg WHO_{2005}-TEQ m^{-2} $year^{-1}$ in 2014, 2015 and 2016. An increase in rainfall may cause a rise in wet deposition, and the annual rainfall was 930.6, 1288.8 and 1328 mm in 2014, 2015 and 2016. In total, the annual total depositions (dry deposition + wet deposition) were 6,010, 6,110 and 5,740 pg WHO_{2005}-TEQ m^{-2} $year^{-1}$ in 2014, 2015 and 2016, respectively. During 2014, 2015 and 2016, the annual average dry, wet and total depositions were 5,510, 439 and 5,950 pg WHO_{2005}-TEQ m^{-2} $year^{-1}$, and the contribution of dry deposition to total deposition was 92.6%.

Table 5. Observed atmospheric PCDD/Fs concentration in Yunlin in 2014.

	6–9 January, 2014					
	QT	ML1	ML2	TX1	TX2	TX3
2,3,7,8-TeCDD	0.0033	0.00447	0.00488	0.00368	0.00351	0.00219
1,2,3,7,8-PeCDD	0.0104	0.015	0.0171	0.00891	0.0112	0.00576
1,2,3,4,7,8-HxCDD	0.0112	0.0145	0.0181	0.00883	0.0105	0.00535
1,2,3,6,7,8-HxCDD	0.0223	0.0314	0.0399	0.0184	0.0221	0.0109
1,2,3,7,8,9-HxCDD	0.0171	0.0237	0.0305	0.0135	0.0181	0.00838
1,2,3,4,6,7,8-HpCDD	0.126	0.19	0.292	0.103	0.12	0.082
OCDD	0.204	0.446	0.8	0.199	0.202	0.35
2,3,7,8-TeCDF	0.0342	0.0431	0.0406	0.0251	0.0338	0.0231
1,2,3,7,8-PeCDF	0.0478	0.0618	0.0745	0.0355	0.0458	0.0318
2,3,4,7,8-PeCDF	0.0748	0.0988	0.134	0.0523	0.0699	0.0462
1,2,3,4,7,8-HxCDF	0.0854	0.132	0.192	0.0718	0.078	0.0584
1,2,3,6,7,8-HxCDF	0.0775	0.112	0.162	0.0614	0.073	0.0492
2,3,4,6,7,8-HxCDF	0.102	0.161	0.261	0.082	0.0881	0.0641
1,2,3,7,8,9-HxCDF	0.00829	0.011	0.0112	0.00614	0.00564	0.00539
1,2,3,4,6,7,8-HpCDF	0.27	0.671	1.22	0.328	0.263	0.232
1,2,3,4,7,8,9-HpCDF	0.0411	0.0899	0.166	0.0393	0.034	0.0366
OCDF	0.184	0.925	1.8	0.402	0.219	0.313
PCDDs (pg m^{-3})	0.395	0.725	1.2	0.355	0.387	0.464
PCDFs (pg m^{-3})	0.925	2.3	4.06	1.1	0.91	0.859
PCDDs/PCDFs	0.427	0.315	0.296	0.322	0.425	0.541
PCDD/Fs (pg m^{-3})	1.32	3.03	5.26	1.46	1.3	1.32
PCDDs (pg WHO_{2005}-TEQ m^{-3})	0.0201	0.0285	0.0340	0.0178	0.0210	0.0113
PCDFs (pg WHO_{2005}-TEQ m^{-3})	0.0578	0.0853	0.1235	0.0452	0.0532	0.0376
PCDDs/PCDFs (WHO_{2005}-TEQ)	0.348	0.334	0.275	0.393	0.395	0.301
PCDD/Fs(pg WHO_{2005}-TEQ m^{-3})	0.0779	0.1138	0.1575	0.0629	0.0743	0.0490
	12–15 August, 2014					
	QT	ML1	ML2	TX1	TX2	TX3
2,3,7,8-TeCDD	0.00076	0.000696	0.000842	0.000826	0.00068	0.000964
1,2,3,7,8-PeCDD	0.000875	0.000768	0.000835	0.00092	0.00124	0.000797
1,2,3,4,7,8-HxCDD	0.000675	0.000757	0.000893	0.000628	0.00071	0.000515
1,2,3,6,7,8-HxCDD	0.00154	0.00124	0.00162	0.0012	0.00145	0.00134
1,2,3,7,8,9-HxCDD	0.000935	0.000771	0.00111	0.000972	0.00101	0.000678
1,2,3,4,6,7,8-HpCDD	0.0104	0.00995	0.011	0.009	0.012	0.00497
OCDD	0.0525	0.06253	0.0529	0.0583	0.0774	0.0261
2,3,7,8-TeCDF	0.00368	0.00425	0.00425	0.00405	0.00453	0.00374
1,2,3,7,8-PeCDF	0.00391	0.00449	0.00427	0.00408	0.00423	0.00407
2,3,4,7,8-PeCDF	0.00455	0.00476	0.00449	0.00433	0.00482	0.00436
1,2,3,4,7,8-HxCDF	0.00389	0.00449	0.00399	0.00386	0.00425	0.00361
1,2,3,6,7,8-HxCDF	0.00423	0.00376	0.00427	0.00367	0.00452	0.0036
2,3,4,6,7,8-HxCDF	0.00366	0.00283	0.00316	0.00306	0.00353	0.00273
1,2,3,7,8,9-HxCDF	0.000616	0.000564	0.000667	0.000863	0.000587	0.000845
1,2,3,4,6,7,8-HpCDF	0.0157	0.0155	0.0158	0.0137	0.01929	0.00805
1,2,3,4,7,8,9-HpCDF	0.00174	0.00158	0.00172	0.00134	0.00156	0.00124
OCDF	0.069	0.0719	0.0602	0.0627	0.135	0.0254
PCDDs (pg m^{-3})	0.0676	0.0767	0.0692	0.0719	0.0945	0.0354
PCDFs (pg m^{-3})	0.111	0.114	0.103	0.102	0.183	0.0576
PCDDs/PCDFs	0.61	0.672	0.673	0.708	0.518	0.614
PCDD/Fs (pg m^{-3})	0.179	0.191	0.172	0.173	0.277	0.093
PCDDs (pg WHO_{2005}-TEQ m^{-3})	0.0021	0.0019	0.0022	0.0021	0.0024	0.0021
PCDFs (pg WHO_{2005}-TEQ m^{-3})	0.0033	0.0033	0.0033	0.0031	0.0036	0.0030
PCDDs/PCDFs (WHO_{2005}-TEQ)	0.630	0.556	0.656	0.679	0.668	0.695
PCDD/Fs(pg WHO_{2005}-TEQ m^{-3})	0.0054	0.0052	0.0055	0.0053	0.0059	0.0051

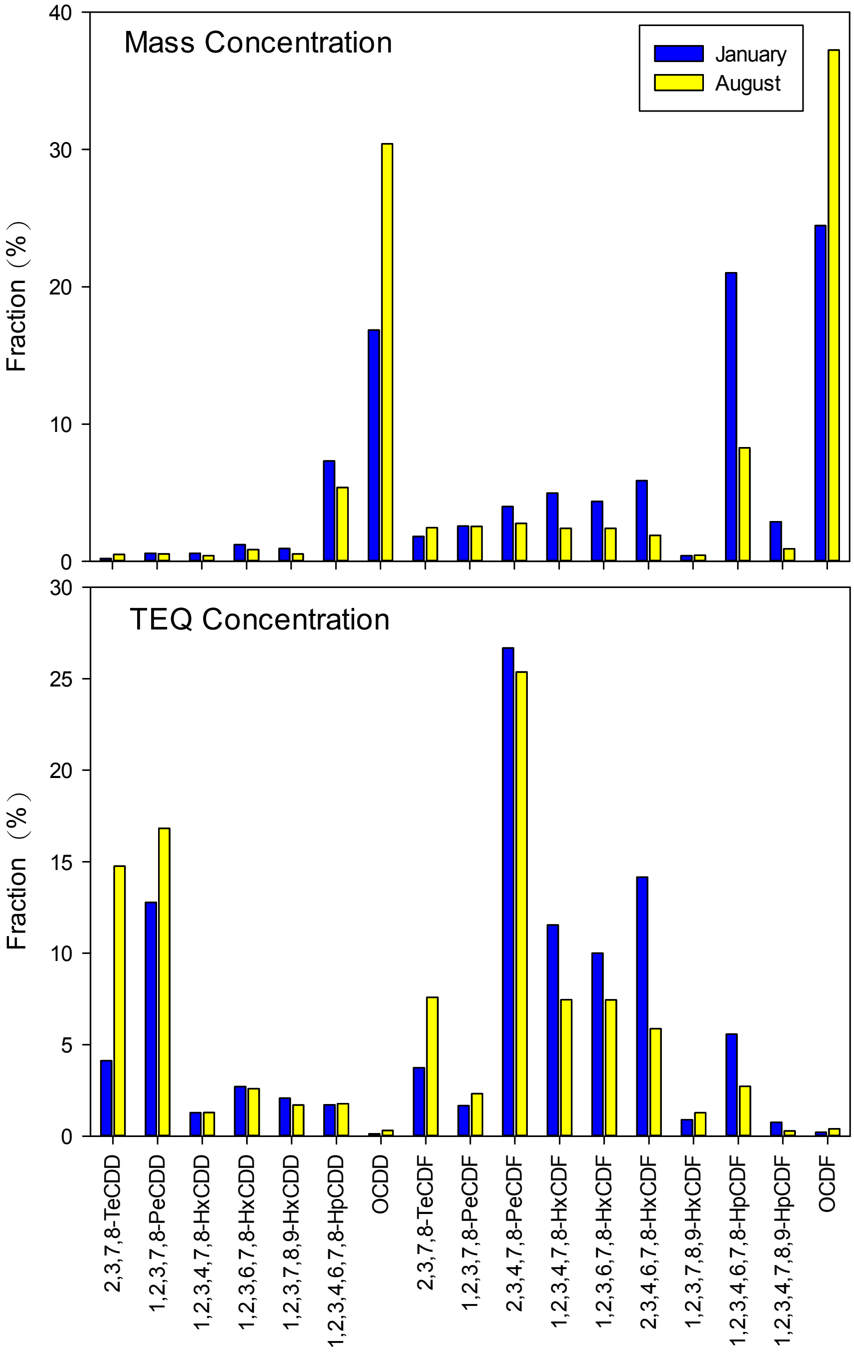

Fig. 2. Congener profiles of PCDD/F mass and TEQ fractions in the ambient Air of Yunlin.

As for Taisi, the annual dry deposition fluxes of total-PCDD/Fs-WHO_{2005}-TEQ were 4,230, 3,720 and 3,580 pg WHO_{2005}-TEQ m^{-2} $year^{-1}$ in 2014, 2015 and 2016, respectively, and the annual wet deposition fluxes of total-PCDD/Fs-WHO_{2005}-TEQ were 251, 313 and 548 pg WHO_{2005}-TEQ m^{-2} $year^{-1}$ in 2014, 2015 and 2016, respectively. As noted above, an increase in rainfall may cause an increase in wet deposition, and the annual rainfall was 1,342, 1,712.2 and 1,862.2 mm in 2014, 2015 and 2016. Overall, the annual total deposition fluxes of total-PCDD/Fs-WHO_{2005}-TEQ were 4,480, 4,030 and 4,130 pg WHO_{2005}-TEQ m^{-2} $year^{-1}$ in 2014, 2015 and 2016, respectively. In 2014, 2015 and 2016, the annual average dry, wet and total deposition fluxes were 3,840, 371 and 4,210 pg WHO_{2005}-TEQ m^{-2} $year^{-1}$, and the contribution of dry deposition to total deposition was 91.2%.

The results indicate that the dry deposition decreased year by year in all three areas, and this may be due to the decrease in PM_{10} concentrations in the ambient air from 2014 to 2015. In contrast, the wet deposition fluctuated because the rainfall varied. Similar with previous studies, the dry and total deposition show seasonal variations in the three areas (Wu *et al.*, 2009; Wang *et al.*, 2010; Mi *et al.*, 2012). From 2014 to 2016, the annual average dry, wet and total

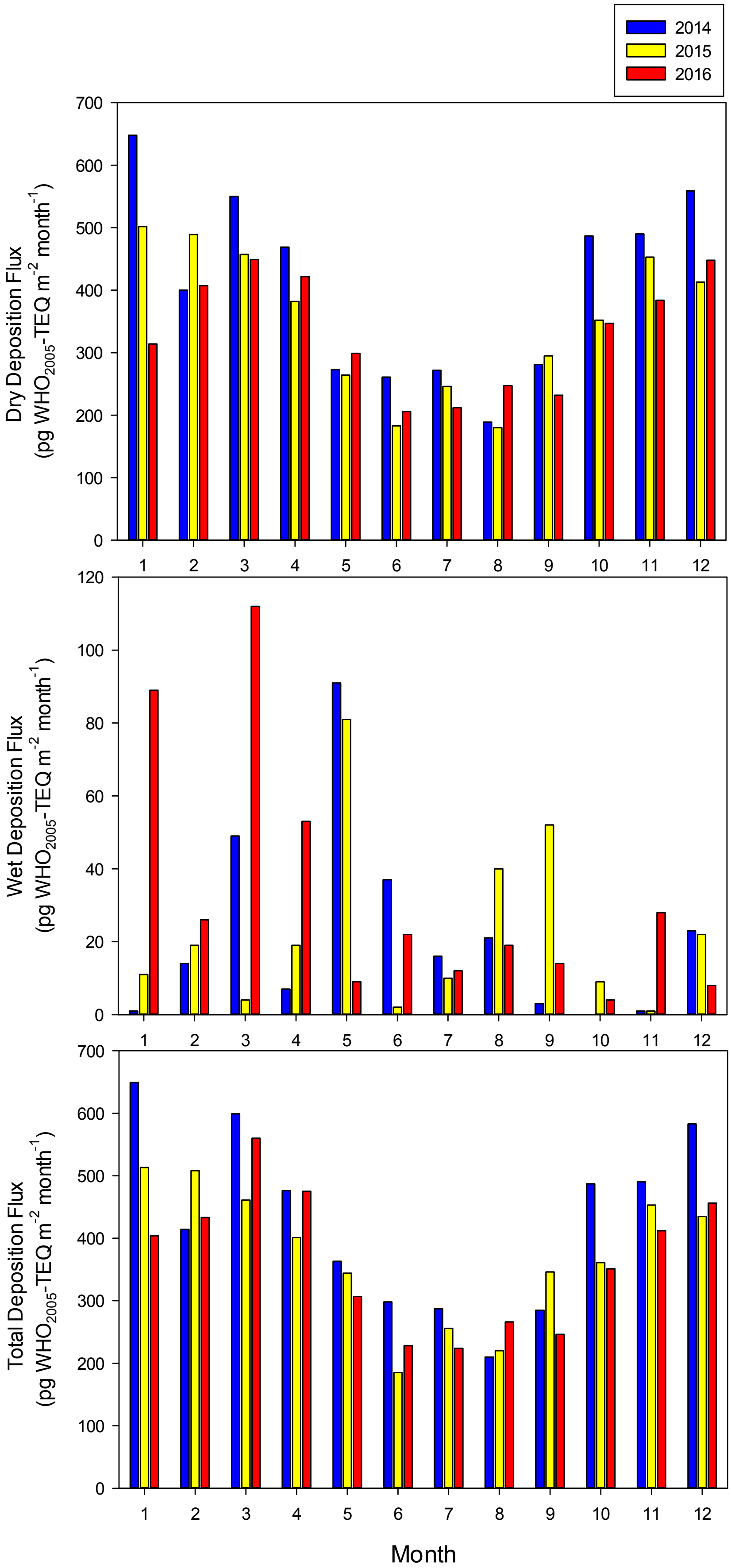

Fig. 3. Monthly average dry deposition, wet deposition and total deposition flux of PCDD/Fs in Lunbei from 2014 to 2016.

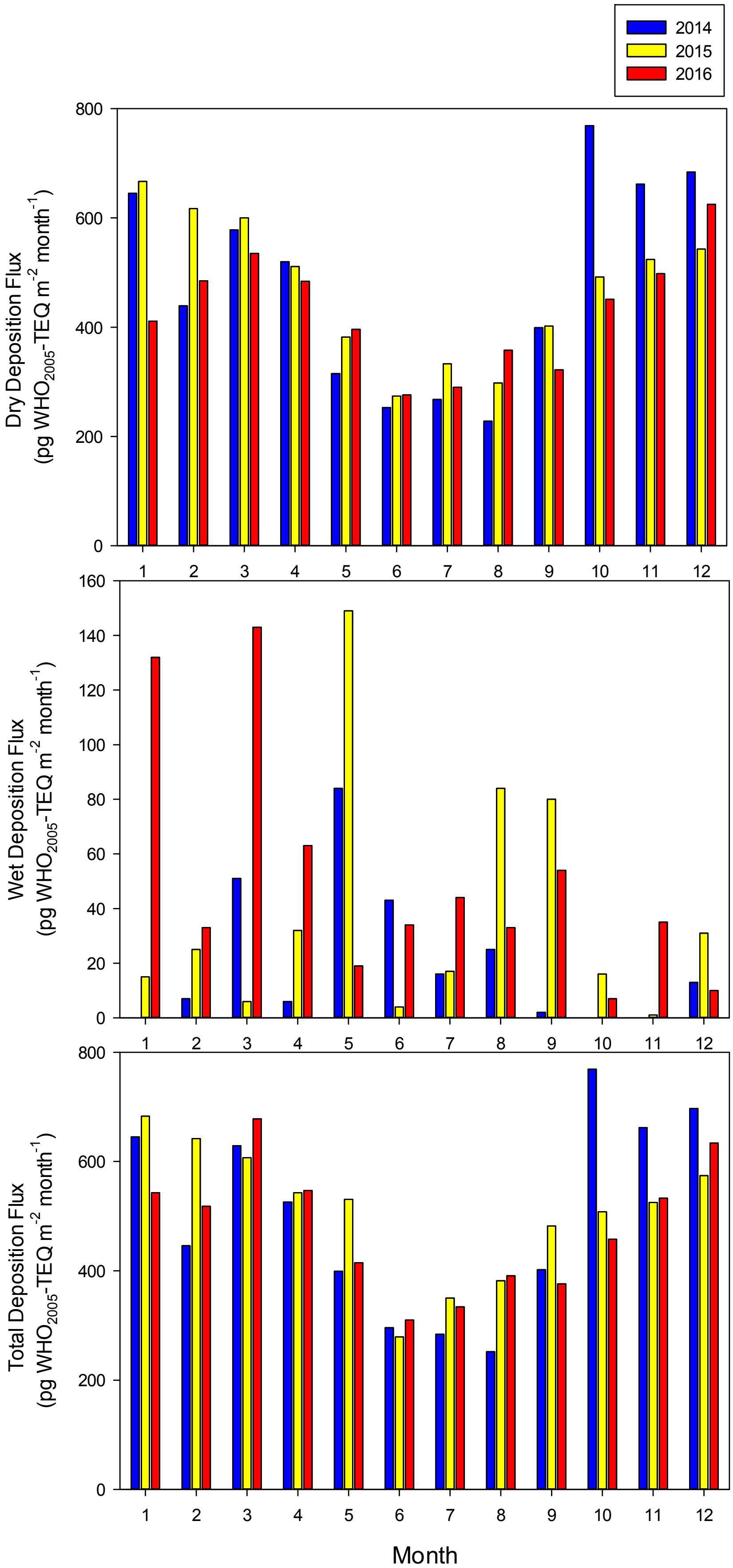

Fig. 4. Monthly average dry deposition, wet deposition and total deposition flux of PCDD/Fs in Mailiao from 2014 to 2016.

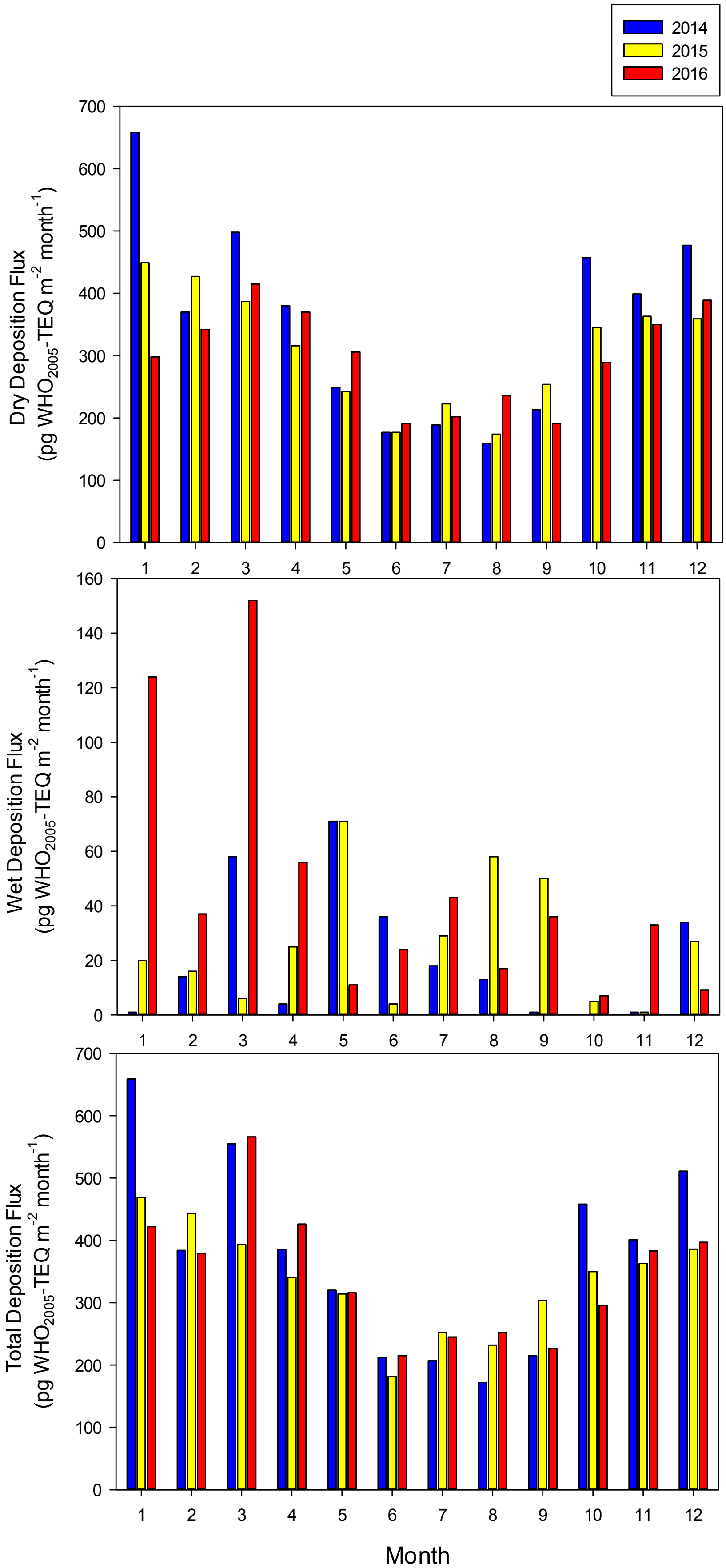

Fig. 5. Monthly average dry deposition, wet deposition and total deposition flux of PCDD/Fs in Taisi from 2014 to 2016.

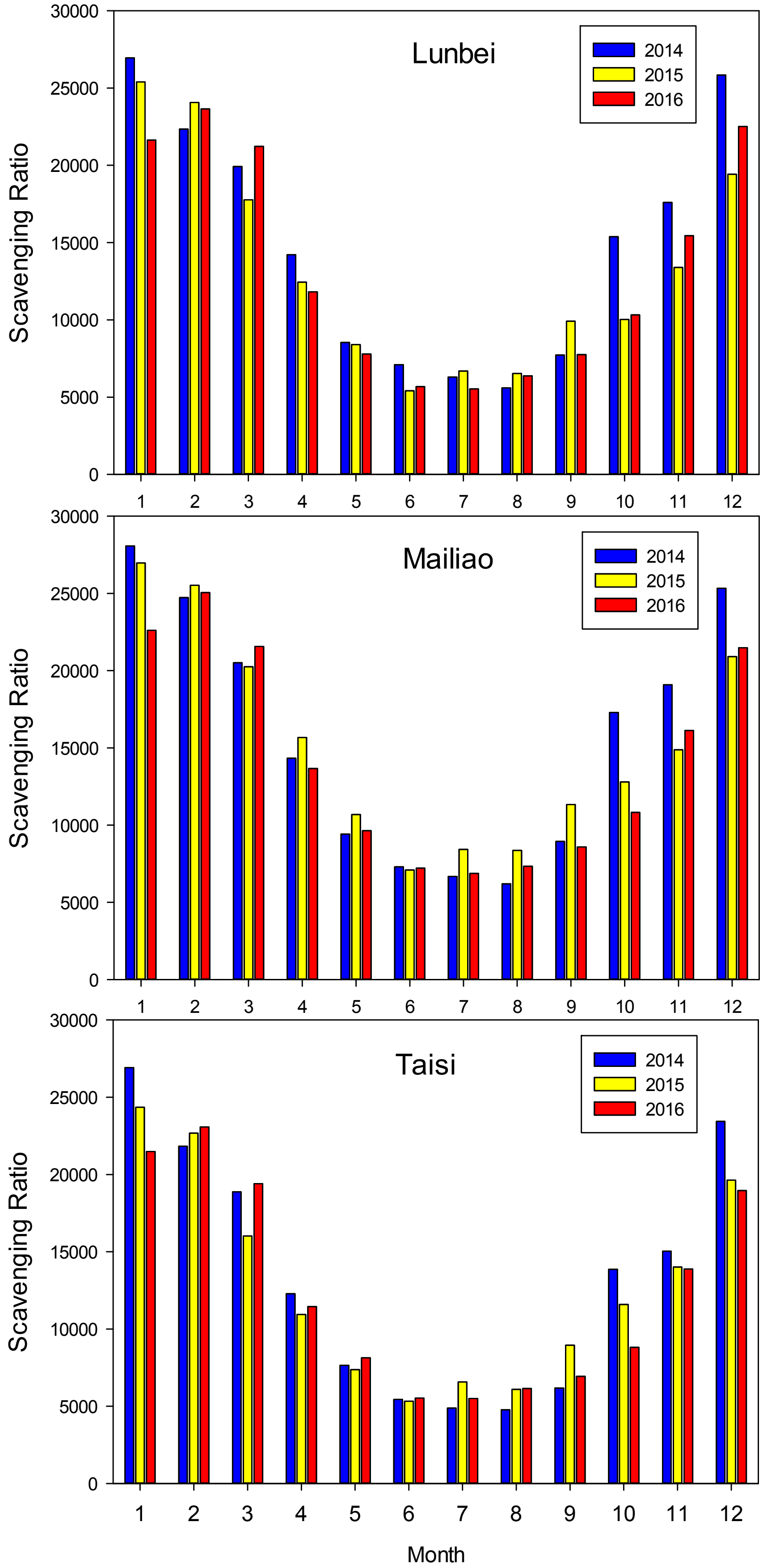

Fig. 6. Monthly average scavenging ratio of total-PCDD/Fs-WHO_{2005}-TEQ in Yunlin from 2014 to 2016.

deposition in Yunlin were 4,960, 254 and 5,210 pg WHO_{2005}-TEQ m^{-2} $year^{-1}$ in 2014; 4,520, 348 and 4,870 pg WHO_{2005}-TEQ m^{-2} $year^{-1}$ in 2015; and 4,220, 518 and 4,740 pg WHO_{2005}-TEQ m^{-2} $year^{-1}$ in 2016. The total deposition of PCDD/Fs in this study was similar with that of rural area in Taiwan and Lagoon in France (Guerzoni *et al.*, 2004; Huang *et al.*, 2011). Among the three areas examined in this work, the dry deposition fluxes were highest at Mailiao, followed by Lunbei, and then Taisi.

The total scavenging ratios (S_{tot}) increased with a decrease of temperature in the three townships from 2014 to 2016, and the results are presented in Fig. 6. In Lunbei, the S_{tot} of total-PCDD/Fs-WHO_{2005}-TEQ were in the ranges of 5,590–27,000, 5,410–25,400, 5,530–23,700, with an average of 14,800, 13,300, 13,300 in 2014, 2015 and 2016, respectively. While in Mailiao, the scavenging ratios (S_{tot}) of total-PCDD/Fs-WHO_{2005}-TEQ were in the ranges of 6,190–28,100, 7,090–27,000, and 6,870–25,000 and with an average of 15,700, 15,200, and 14,200 in 2014, 2015 and 2016, respectively. In Taisi, the scavenging ratios (S_{tot}) of total-PCDD/Fs-WHO_{2005}-TEQ were in the ranges of 4,780–26,900, 5,330–24,300, 5,500–23,100 and with an average of 13,400, 12,800, and 12,400 in 2014, 2015 and 2016, respectively. From 2014 to 2016, the mean scavenging ratios (S_{tot}) of total-PCDD/Fs-WHO_{2005}-TEQ were 13,800, 15,000 and 12,900 in Lunbei, Mailiao and Taisi, respectively. As the results show, the scavenging ratios (S_{tot}) indicate a distinctive seasonal variation. From 2014 to 2016, the minimum values of S_{tot} occur in summer, while the maximum occur in winter, the S_{tot} in spring, summer, autumn and winter were 13,700, 6,330, 12,100 and 23,500 in Yunlin, with an average temperature of 23.4, 28.7, 26.1 and 17.8°C, respectively. This is due to the fact that a higher temperature will cause a greater fraction of PCDD/Fs in the gas phase in summer and the gas phase scavenging ratio is less than that of the particle phase. Overall, the scavenging ratios of total-PCDD/Fs-WHO_{2005}-TEQ in Yunlin were 14,600, 13,800 and 13,300 in 2014, 2015 and 2016, respectively, with an average of 13,900. The mean S_{tot} in Yunlin (13,900) is similar to the value in Lulin (13,450), in 2012 and 2013 (Chandra *et al.*, 2015), but higher than in Xiaogang (6,840) and Meinong (4,700) (Lee *et al.*, 2016).

CONCLUSION

1. The mean $PM_{2.5}$ concentrations in Yunlin were 29.6, 26.6 and 26.3 µg m^{-3} in 2014, 2015 and 2016, respectively. The mean values of $PM_{2.5}/PM_{10}$ ratio are in an order of Lunbei (averaged at 0.553) > Taisi (averaged at 0.536) > Mailiao (averaged at 0.363). It is more dangerous for human health in Lunbei and Taisi than Mailiao.
2. The concentration of total-PCDD/Fs-WHO_{2005}-TEQ decreased year after year from 2014 to 2016. The mean PCDD/Fs concentrations at Yunlin were 0.038, 0.0346 and 0.0324 pg WHO_{2005}-TEQ m^{-3} from 2014 to 2016.
3. In 2014, the mass concentrations of total PCDD/Fs in January were much higher than those in August at all sample sites. In January 2014, the PCDD/Fs concentrations in QT, ML1, ML2, TX1, TX2 and TX3 were 1.32, 3.03, 5.26, 1.46, 1.3 and 1.32 pg m^{-3}, respectively; while in August, the PCDD/Fs concentrations were 1.32, 3.03, 5.26, 1.46, 1.3 and 1.32 pg m^{-3}. The PCDD/F congener profiles of the ambient air samples are largely dominated by higher chlorinated PCDD/F congeners.
4. The observed concentrations of total-PCDD/Fs-WHO_{2005}-TEQ at Yunlin in January and August were 0.089 and 0.00538 pg WHO_{2005}-TEQ m^{-3}, and the modeled concentrations were 0.0598 and 0.0176 pg WHO_{2005}-TEQ m^{-3}.
5. From 2014 to 2016, the annual average dry, wet and total deposition in Yunlin were 4,955, 254 and 5,209 pg WHO_{2005}-TEQ m^{-2} $year^{-1}$ in 2014; 4,524, 348 and 4,872 pg WHO_{2005}-TEQ m^{-2} $year^{-1}$ in 2015; 4,224, 518 and 4,742 pg WHO_{2005}-TEQ m^{-2} $year^{-1}$ in 2016. Among the three areas, the dry deposition fluxes are highest in Mailiao, followed by Lunbei, and Taisi.
6. The scavenging ratios (S_{tot}) show a distinctive seasonal variation, and the values in spring, summer, autumn and winter were 13,700, 6,330, 12,100 and 23,500 in Yunlin, respectively. This is due to the fact that a higher temperature will cause a greater fraction of PCDD/Fs in the gas phase in summer and the gas phase scavenging ratio is less than that of the particle phase.
7. The S_{tot} of total-PCDD/Fs-WHO_{2005}-TEQ in Yunlin were 14,600, 13,800 and 13,300 in 2014, 2015 and 2016, respectively, with an average of 13,900.
8. The results of this study provide information show the trends of both atmospheric $PM_{2.5}$ and PCDD/Fs in central Taiwan. This data is useful for the establishment of control strategies in future works.

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