Aerosol and Air Quality Research, 17: 1798–1810, 2017

ISSN: 1680-8584 print / 2071-1409 online
doi: 10.4209/aaqr.2017.05.0177

Atmospheric Deposition of Polychlorinated Dibenzo-*p*-Dioxins and Dibenzofurans in Two Cities of Southern China

Jinning Zhu[1], Haiyan Tang[1], Jin Xing[1], Wen-Jhy Lee[1,2*], Ping Yan[1], Kangping Cui[1***]**

[1] *School of Resources and Environmental Engineering, Hefei University of Technology, Hefei 246011, China*
[2] *Department of Environmental Engineering, National Cheng Kung University, Tainan 70101, Taiwan*

ABSTRACT

Atmospheric deposition is an important pathway for air pollutants entering the environment. In order to better understand both dry and wet deposition of PCDD/Fs in ambient air, two cities – Guangzhou and Nanjing in Southern China, were investigated. The monthly dry deposition fluxes of total-PCDD/Fs-WHO_{2005}-TEQ were in the range of 60.6–560 and 104–1160 pg WHO_{2005}-TEQ m^{-2} $month^{-1}$ during 2014 in Guangzhou and Nanjing, respectively. In addition, it was found that the monthly dry deposition velocities of particle phase PCDD/Fs-TEQ ranged between 0.49 and 0.98 cm s^{-1} (averaged 0.69 cm s^{-1}) and between 0.44 and 0.8 cm s^{-1} (averaged 0.52 cm s^{-1}) in Guangzhou and Nanjing, respectively. The average scavenging ratios of total-PCDD/Fs-WHO_{2005}-TEQ were 20480 and 30947 in Guangzhou and Nanjing, respectively. The total (dry + wet) deposition fluxes in Nanjing ranged between 135 and 1250, and averaged 643 pg WHO_{2005}-TEQ m^{-2} $month^{-1}$, approximatedly1.38–2.23 times of magnitude higher than those in Guangzhou, which ranged between 97.7 to 559 pg WHO_{2005}-TEQ m^{-2} $month^{-1}$ and averaged 254 pg WHO_{2005}-TEQ m^{-2} $month^{-1}$. The results of this study provide useful information for both further studies and environmental control strategies aimed at persistent organic compounds (POPs).

Keywords: Dry deposition; Wet deposition; PCDD/Fs; PM_{10}; $PM_{2.5}$; Southern China.

INTRODUCTION

Increased vehicular traffic and other combustion processes have resulted in a significant increase in ambient particulate matter (PM) over the past two decades. Fine particulate matter (dp ≤ 2.5 μm, $PM_{2.5}$) has aroused widespread public concern, since it has been found to contain toxic compounds like PCDD/Fs, contribute to poor air quality, and have adverse effects on human health and global climate change (Laden *et al*., 2000; Chow *et al*., 2015). Previous investigations have shown good correlations between PM_{10} and total-PCDD/Fs mass concentrations (Wang *et al*., 2010; Lee *et al*., 2016). Higher $PM_{2.5}$ and PCDD/Fs levels are contributed by anthropogenic rather than natural sources, with seasonal variations in these (Yu *et al*., 2013; Wang *et al*., 2015; Lu *et al*., 2016). Ambient particulate matters can be removed from atmosphere by both dry and wet deposition (Cheruiyot *et al*., 2015, 2016).

Polychlorinated dibenzo-*p*-dioxins (PCDDs) and polychlorinated dibenzofurans (PCDFs) are well known as persistent organic pollutants (POPs), which are formed during any incomplete combustion processes (Bumb *et al*., 1980). Human activities, including industrial and heat-treatment processes, such as municipal solid waste incinerators (MSWIs), medical waste incinerators, and hazardous waste incinerators (HWIs), are the major source of PCDD/F emissions to the atmosphere (Oh *et al*., 1999; Addink and Altwicker, 2001; Neuer-Etscheidt *et al*., 2006; Lin *et al*., 2014; Cheruiyot *et al*., 2016), and also occur in natural combustion processes such as volcanic eruptions and forest fires (Prange *et al*., 2002; Kim *et al*., 2003; Prange *et al*., 2003). There are 210 possible congeners of PCDD/Fs (75 PCDDs and 135 PCDFs), and of these 17 with chlorine atoms attached to the 2, 3, 7 and 8 positions have been shown to be more toxic, with highly adverse effects on human health. The major distribution pathway of PCDD/Fs is through air (Lohmann and Jones, 1998; Kao *et al*., 2006; Lee *et al*., 2009), and they can remain in the environment for a long time and transport long distances (Lee *et al*, 2003; Zhang *et al*., 2009), resulting in the widespread distribution of

* Corresponding author.
Tel.: +886-913-027-189
E-mail address: lwjlee@mail.ncku.edu.tw
** Corresponding author.
Tel.: +86-188-0551-2007
E-mail address: y9621227@hfut.edu.cn
*** Corresponding author.
Tel.: +86-186-5595-3355
E-mail address: cuikangping@163.com

PCDD/Fs through the environment and accumulation in the soil, water or food (Lohmann and Jones, 1998; Hu *et al*., 2009).

Previous studies have indicated that the factors of ambient temperature, humidity, vapor pressure and the chemical properties of compounds have important influences on the partition fractions between the gas and particulate phases (Lohmann *et al*., 1999; Chang *et al*., 2004; Lee *et al*., 2008; Wang *et al*., 2010; Lee *et al*., 2016), and thus can cause significant differences in atmospheric deposition. For example, the gas-phase decreases as the chlorination level of PCDD/Fs increases, while a 10°C decrease in the ambient air temperature will cause a 20% increase of the PCDD/Fs bound to particulate phase (Chang *et al*., 2004). The atmospheric deposition of PCDD/Fs can be divided into dry and wet deposition, respectively. The dry deposition is contributed by both gas and particle phases, while gas phase deposition includes absorption at the air-liquid or air-solid interfaces and depends significantly on the gas-particle partition (Oh *et al*., 2001). Turbulent diffusion, sedimentation, inertial forces, electrical migration and diffusiophoresis are the major methods of dry deposition. However, wet deposition is the removal of atmospheric particles by precipitation such as rain, cloud droplets or snow (Lohmann and Jones, 1998), and it is more relevant in regions with heavy rain seasons, higher precipitation rates and/or higher chlorinated homologues in the environmental sinks (Shih *et al*., 2006; Wang *et al*., 2010).

As the largest developing country in the world, the air quality in China is much worse than the recommended standards set out by the World Health Organization (WHO), posing significant threats to human health. With its rapid economic development, quick industrialization and urbanization, the level of air pollution in China has attracted global attention (Xu *et al*., 2013). Research shows that half of the MSWIs in China exceed the national standard of 1.0 ng I-TEQ Nm^{-3} for PCDD/Fs emissions (Tian and Ouyang, 2003), and only two of the 14 domestically made medical waste incinerators are under the European Union standards of 0.1 ng I-TEQ Nm^{-3} in China (Gao *et al*., 2009). The PCDD/Fs concentrations are in the range of 51.2–61.9 fg WHO-TEQ m^{-3} in the air near MSWIs in China (Meng *et al*., 2016). Dry and wet deposition are effective and important mechanisms for the removal of atmospheric pollutants. Although some PCDD/F emission data and that on the deposition of atmospheric pollutants has been reported (Liu *et al*., 2006; Guo *et al*., 2014; Wang *et al*., 2014; Chandra Suryani *et al*., 2015), there is still lack of studies on the dry and wet deposition of PCDD/Fs in China. Therefore, this study aims to investigate the atmospheric deposition fluxes of PCDD/Fs in mainland China. The results from this study provide a means to further identify the environmental characteristics associated with the concentration and atmospheric deposition of PCDD/Fs in ambient air.

During 2014, this study examined the atmospheric dry, wet and total (dry + wet) deposition fluxes of PCDD/Fs in two typical cities (Nanjing and Guangzhou) in Southern China. In addition, for PCDD/Fs-WHO_{2005}-TEQ, the dry deposition velocities of the particle phase and wet deposition scavenging ratios in two typical southern cities (Nanjing and Guangzhou) were studied. Furthermore, the monthly, seasonal and annual variations in PCDD/Fs depositions were compared and discussed.

METHODS

Sample Sites

The monthly mean concentrations and gas-particle distributions of ambient PCDD/Fs-WHO_{2005}-TEQ from January 2014 to December 2014 in Southern China were obtained from Tang *et al*. (2017). In this study, two typical cities were selected and evaluated, which are Nanjing in Jiangsu province, and Guangzhou in Guangdong province. The pertinent meteorological information, included monthly temperature and rainfall for these two areas in 2014, was obtained from local air quality monitoring stations.

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:

$$F_{d,T} = F_{d,g} + F_{d,p}$$
$$C_T \times V_{d,T} = C_g \times V_{d,g} + C_p \times V_{d,p} \quad (1)$$

$F_{d,T}$: 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). Dry deposition of gas-phase PCDD/Fs is mainly by diffusion, and due to the lack of measured data for PCDD/Fs, a selected value (0.010 cm s^{-1}) for the gas-phase PAH dry deposition velocity, $V_{d,g}$, as proposed by Sheu *et al.* (1996) and used by Lee *et al.* (1996), is used in the current work 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. (1).

Scavenging Ratios

For a slightly soluble trace organic compound, such as PCDD/Fs, it is commonly believed that equilibrium partitioning occurs between the compound in the gas phase and that in 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:

$$S_g = RT/H \quad (2)$$

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 the ratio of the concentration of the dissolved phase in the raindrop divided by the concentration of the gas phase in the air, S_g, and can be calculated by:

$$S_g = C_{rain,dis}/C_g \quad (3)$$

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 the ratio of the concentration of the particle phase in the raindrop divided by the concentration of the particle phase in the air, S_p, which can be calculated by:

$$S_p = C_{rain,particle}/C_p \quad (4)$$

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.

The total scavenging of precipitation is the sum of gas and particle scavenging, S_{tot}, which can be calculated by:

$$S_{tot} = S_g(1 - \Phi) + S_p \times \Phi \quad (5)$$

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

Because of a lack of measured data for the particle scavenging ratios of PCDD/Fs, the S_p (S_p is 42,000), the 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, 2011b). 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:

$$F_{w,T} = F_{w,dis} + F_{w,p} \quad (6)$$

$$F_{w,dis} = C_{rain,dis} \times Rainfall \quad (7)$$

$$F_{w,p} = C_{rain,particle} \times Rainfall \quad (8)$$

$F_{w,T}$: 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

In this study, the monthly concentrations of total-PCDD/Fs-WHO_{2005}-TEQ in the ambient air are derived from the particulate matter ($PM_{2.5}$, PM_{10}, TSP). The gas-particle phase distributions and the concentrations of total-PCDD/Fs-WHO_{2005}-TEQ in the ambient air during 2014 in both cities, as reported in Tang *et al*. (2017).

Dry Deposition

Based on deposition Eq. (1), the monthly dry deposition fluxes of total-PCDD/Fs-WHO_{2005}-TEQ for both cities during 2014 are calculated and shown in Fig. 1. C_T was modeled and input into the Eq. (1), while C_p and C_g were calculated by gas/particle partitioning. Based on these parameters, F_g ang F_p were determined by the deposition velocity.

During 2014, in Guangzhou City, the monthly dry deposition fluxes of total-PCDD/Fs-WHO_{2005}-TEQ ranged between 60.6 and 558 pg WHO_{2005}-TEQ m^{-2} $month^{-1}$, with averages of 186, 64.3, 177 and 396 pg WHO_{2005}-TEQ m^{-2} $month^{-1}$ in spring, summer, fall and winter, respectively. In total, the annual total dry deposition fluxes of total PCDD/Fs-WHO_{2005}-TEQ was 2470 pg WHO_{2005}-TEQ m^{-2} $year^{-1}$. Compared with previous studies, our results for the monthly total dry deposition fluxes of PCDD/Fs in Guangzhou (2.02–18.6 pg WHO_{2005}-TEQ m^{-2} day^{-1}) were very similar with those seen in rural areas in Taiwan (3.07–18.9 pg WHO_{2005}-TEQ m^{-2} day^{-1}) (Shih *et al*., 2006). Because the PCDD/Fs are mainly in the particle-phase in the ambient air, our results demonstrated that the particle-bond deposition fluxes of PCDD/Fs (84.9%–95.9%) contributed far more than gas phase deposition fluxes to the total dry deposition fluxes, and that the monthly dry deposition fluxes of PCDD/Fs contributed by the gas and particle phases were 9.08–22.7 (lowest in August and highest in January) and 51.5–535 (lowest in August and highest in January) pg WHO_{2005}-TEQ m^{-2} $month^{-1}$, respectively. The monthly average temperature in Guangzhou ranged from 13°C (January and February) to 28.9°C (July), and the annual average temperature was 21.7°C. As in previous studies (Sugita *et al*., 1994; Shih *et al*., 2006; Huang *et al*., 2011a), the estimated monthly fluctuations of the dry deposition fluxes of PCDD/Fs suggest that these reached the highest level in the cold season (January) and the lowest in the warm season (August).

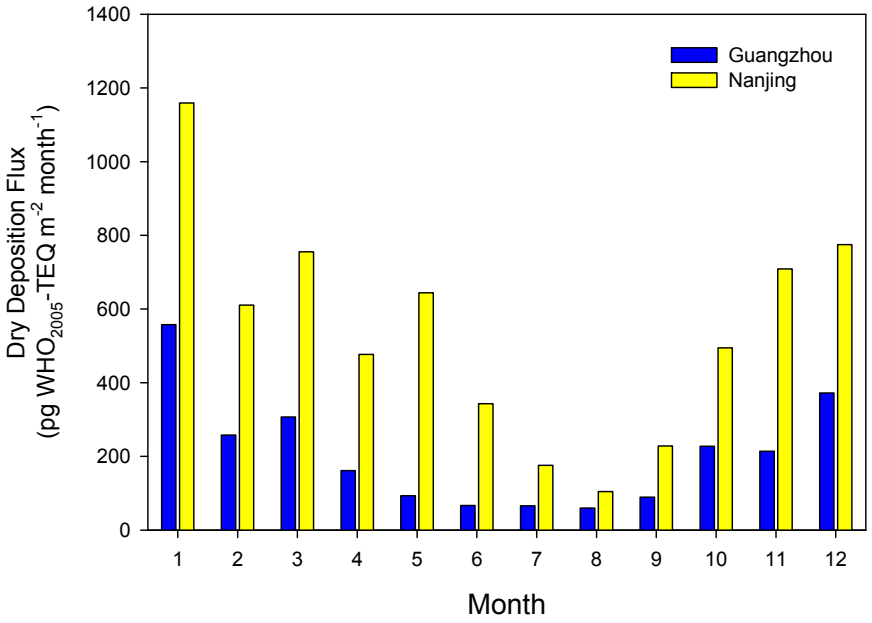

Fig. 1. Monthly Average Dry Deposition Flux of total-PCDD/Fs-WHO_{2005}-TEQ during 2014.

However, in Nanjing City, the results indicated that the monthly dry deposition fluxes of total-PCDD/Fs-WHO_{2005}-TEQ were approximately twice of Guangzhou during 2014, and ranged between 104 and 1160 pg WHO_{2005}-TEQ m^{-2} $month^{-1}$, and the annual total dry deposition flux of total-PCDD/Fs-WHO_{2005}-TEQ was 6480 pg WHO_{2005}-TEQ m^{-2} $year^{-1}$. A lower average temperature and a higher atmospheric $PM_{2.5}$ concentration did result in a higher dry deposition flux in Nanjing than in Guangzhou City. The seasonal variations of Nanjing City shown averages of 625, 208, 477 and 848 pg WHO_{2005}-TEQ m^{-2} $month^{-1}$ in spring, summer, fall and winter, respectively. These values were similar to those in Taiwan (Shih *et al*., 2006; Wu *et al*., 2009; Mi *et al*., 2012). As in with Guangzhou, the total dry deposition fluxes of PCDD/Fs in Taiwan are mainly contributed by the particle-bound deposition fluxes (87.1%–98.8%), and the value reach the highest level in January and the lowest in August. The monthly dry deposition fluxes of PCDD/Fs contributed by the gas phase and particle phase are 10.3–49.0 (lowest in February and highest in May) and 90.9–1150 (lowest in August and highest in January) pg WHO_{2005}-TEQ m^{-2} $month^{-1}$, respectively. The monthly average temperatures in Guangzhou range from 4.7°C (February) to 27.1°C (July), and the average annual temperature is 16.4°C. Comparisons of the dry deposition values found in this and other studies are shown in Table 1.

The dry deposition velocity was determined by the ratio of total dry deposition fluxes and the PCDD/Fs concentrations in the atmosphere. In this study, the dry deposition velocity of total PCDD/Fs ($V_{d,T}$) and the dry deposition velocity of the gas-phase PCDD/Fs ($V_{d,g}$) were selected from previous studies (Lee *et al*., 1996; Sheu *et al*., 1996; Shih *et al*., 2006). Based on Eq. (1), the monthly dry deposition velocities of particle-phase $V_{d,p}$ in both focal cities during 2014 were measured and shown in Fig. 2. The dry deposition velocity of particle-phase total-PCDD/Fs-WHO_{2005}-TEQ ($V_{d,p}$) fluctuated with the temperature, and the velocity reached the highest levels in July (0.98 cm s^{-1} in Guangzhou) and August (0.8 cm s^{-1} in Nanjing), and lowest in January (0.49 cm s^{-1} in Guangzhou, 0.44 cm s^{-1} in Nanjing). These values are similar with to those found for the ambient air near MSWIs in Taiwan (Wu *et al*., 2009; Huang *et al*., 2011a). This is probably due to the fact that in the hot season (for example, July) the mean size of particulate matter (PM) is much higher than that in the cold season (for example, January).

As the results show, in both cities the dry deposition fluxes of total-PCDD/Fs-WHO_{2005}-TEQ were contributed mainly by the particle phase, and the total dry deposition fluxes was found to decrease as the temperature increased. It is probably due to the fact that during the hot season more PCDD/Fs were shifted to the gas phase and there was a higher deposition velocity of particle-phase PCDD/Fs ($V_{d,p}$ ranged between 0.49–0.98 and 0.44–0.8 cm s^{-1}, with an average of 0.69 and 0.52 cm s^{-1} in Guangzhou and Nanjing, respectively), much greater than that of the gas-phase velocity ($V_{d,g}$ was 0.01 cm s^{-1}).

Wet Deposition

The wet deposition fluxes of PCDD/Fs were a combination of both vapor dissolution into rain and the removal of suspended particulate matter by precipitation. Fig. 3 shows the monthly wet deposition flux of total-PCDD/Fs-WHO_{2005}-TEQ in Guangzhou and Nanjing during 2014.

Based on Eqs. (2) and (3), the total PCDD/Fs in the rain's dissolved phase were in the range from 0.0047 to 0.01 pg WHO_{2005}-TEQ L^{-1} in Guangzhou during 2014. In order to estimate the wet deposition fluxes of PCDD/Fs, information on rainfall during 2014 in both cities was obtained from local weather stations. The annual rainfall was 2234 mm in Guangzhou, being highest in May (542.9 mm) and August (513.1 mm). Based on the monthly rainfall and the total-PCDD/Fs-WHO_{2005}-TEQ concentrations in the rain (dissolved and particle phases), the monthly wet deposition fluxes of

total-PCDD/Fs-WHO_{2005}-TEQ were estimated and the values ranged from 0.58 (97.9% contributed by the particle phase) to 229 (99.1% contributed by the particle phase) pg WHO_{2005}-TEQ m^{-2} $month^{-1}$. The results show that the wet deposition fluxes of total-PCDD/Fs-WHO_{2005}-TEQ are dominated by the particle phase, with the same trend present in the dry deposition. The annual total wet deposition fluxes of total-PCDD/Fs-WHO_{2005}-TEQ was 569.6 pg WHO_{2005}-TEQ m^{-2} $year^{-1}$. The highest wet deposition fluxes occurred in March, while the lowest were in October. The seasonal distribution of

Table 1. Comparison of PCDD/Fs-TEQ Dry Deposition Fluxes between Previous Studies and This Study.

Area	Location	Period	Dry Deposition (TEQ)	Reference
Taiwan	Industrial	2010–2011	12.4–20.6 pg I-TEQ m^{-2} day^{-1}	Mi *et al.*, 2012
Taiwan	Urban	2010–2011	13.6–21.8 pg I-TEQ m^{-2} day^{-1}	Mi *et al.*, 2012
Taiwan	Rural	2010–2011	14.3–26.1 pg I-TEQ m^{-2} day^{-1}	Mi *et al.*, 2012
Taiwan	Urban	2010–2011	8.3–13.1 pg I-TEQ m^{-2} day^{-1}	Mi *et al.*, 2012
Taiwan	Urban	2009	3.72–56.8 pg I-TEQ m^{-2} day^{-1}	Wu *et al.*, 2009
Taiwan	Urban	2006	3.07–18.9 pg I-TEQ m^{-2} day^{-1}	Shih *et al.*, 2006
Taiwan	Coastal	2013	33.6–104.3 pg WHO_{2005}-TEQ m^{-2} $month^{-1}$	Chandra Suryani *et al.*, 2015
Taiwan	Mountain	2013	3.9–27.3 pg WHO_{2005}-TEQ m^{-2} $month^{-1}$	Chandra Suryani *et al.*, 2015
Taiwan	Industrial	2014	22.1–632 pg WHO_{2005}-TEQ m^{-2} $month^{-1}$	Lee *et al.*, 2016
Taiwan	Industrial	2015	31.1–468 pg WHO_{2005}-TEQ m^{-2} $month^{-1}$	Lee *et al.*, 2016
Taiwan	Rural	2014	9.7–418 pg WHO_{2005}-TEQ m^{-2} $month^{-1}$	Lee *et al.*, 2016
Taiwan	Rural	2015	7.8–373 pg WHO_{2005}-TEQ m^{-2} $month^{-1}$	Lee *et al.*, 2016
Korea	Urban	2002	2.7–10.1 pg I-TEQ m^{-2} day^{-1}	Moon *et al.*, 2005
Korea	Suburban	2002	3.0–12.6 pg I-TEQ m^{-2} day^{-1}	Moon *et al.*, 2005
USA	Urban	2003–2004	1.5 pg I-TEQ m^{-2} day^{-1}	Correa *et al.*, 2006
Japan	Urban	1996–1998	36–51 pg I-TEQ m^{-2} day^{-1}	Ogura *et al.*, 2001
Italy	Industrial	1998–1999	0–9.2 pg I-TEQ m^{-2} day^{-1}	Guerzoni *et al.*, 2004
Taiwan	Commercial	2006	7.67–18.2 pg I-TEQ m^{-2} day^{-1}	Wang *et al.*, 2010
Taiwan	Industrial	2006	17.6–40.6 pg I-TEQ m^{-2} day^{-1}	Wang *et al.*, 2010
Taiwan	Coastal	2006	6.69–11.6 pg I-TEQ m^{-2} day^{-1}	Wang *et al.*, 2010
Taiwan	Agricultural	2006	5.73–15.6 pg I-TEQ m^{-2} day^{-1}	Wang *et al.*, 2010
Taiwan	Rural	2009–2010	40.6–718 pg I-TEQ m^{-2} $month^{-1}$	Huang *et al.*, 2011a
Taiwan	Rural	2009–2010	27.4–769 pg I-TEQ m^{-2} $month^{-1}$	Huang *et al.*, 2011a
China	Guangzhou	2014	60.6–558 pg WHO_{2005}-TEQ m^{-2} $month^{-1}$	this study
China	Nanjing	2014	104–1160 pg WHO_{2005}-TEQ m^{-2} $month^{-1}$	this study

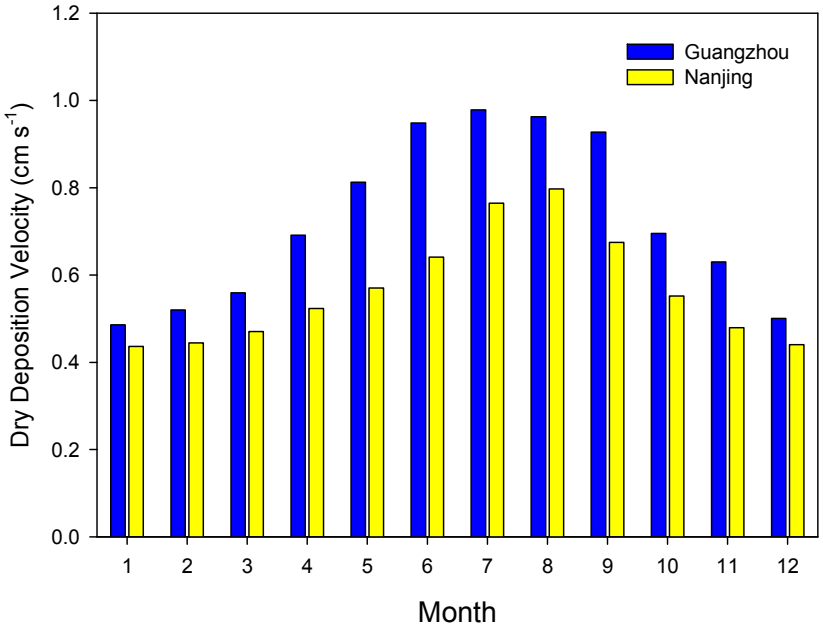

Fig. 2. Monthly Average Dry Deposition Velocity (cm s^{-1}) of Particle-bound total-PCDD/Fs-WHO_{2005}-TEQ during 2014.

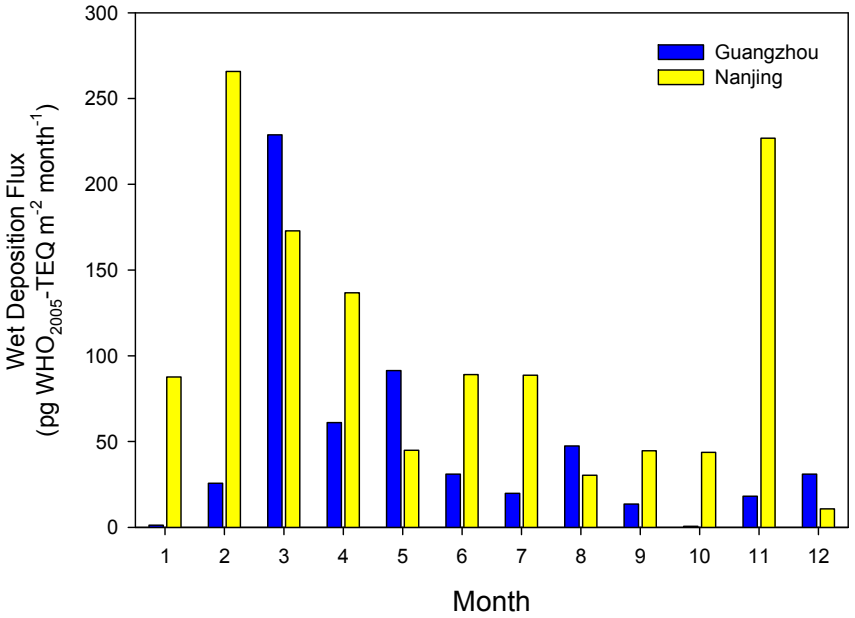

Fig. 3. Monthly Average Wet Deposition Flux of total-PCDD/Fs-WHO_{2005}-TEQ during 2014.

the wet deposition fluxes in Guangzhou were 127, 32.7, 10.8 and 31.3 pg WHO_{2005}-TEQ m^{-2} $month^{-1}$ for spring, summer, autumn and winter, respectively. Comparisons between the wet deposition simulated in this study and in other previous works are shown in Table 2. Based on the monthly wet deposition and rainfall, the concentration of total-PCDD/Fs-WHO_{2005}-TEQ are show in Fig. 4 and ranged between 0.092 (January) to 1.79 (August) pg WHO_{2005}-TEQ L^{-1} in Guangzhou. Comparisons between the concentration in the rain in this study and other previous works are show in Table 3.

However, the total PCDD/Fs-TEQ in the rain's dissolved phase ranged between 0.003 to 0.018 pg total-PCDD/Fs-WHO_{2005}-TEQ L^{-1} in Nanjing during 2014. The annual rainfall was 1091.1 mm in Nanjing, and was strongest in July (263.5 mm) and August (158.8 mm) during 2014. The monthly wet deposition fluxes of total-PCDD/Fs-WHO_{2005}-TEQ ranged from 10.7 (99.9% contributed by the particle phase) to 266 (99.1% contributed by the particle phase) pg total-PCDD/Fs-WHO_{2005}-TEQ m^{-2} $month^{-1}$. The highest wet deposition fluxes occurred in February, while the lowest in December. The average wet fluxes of total-PCDD/Fs-WHO_{2005}-TEQ were 118, 69.4, 105 and 121 pg total-PCDD/Fs-WHO_{2005}-TEQ m^{-2} $month^{-1}$ for spring, summer, autumn and winter, respectively. Although the rainfall in Nanjing was less than that in Guangzhou, the monthly concentrations of total-PCDD/Fs-WHO_{2005}-TEQ in the rain were higher, with a range between 1.91 (January) to 4.26 (August) pg WHO_{2005}-TEQ L^{-1}.

The results show that the wet deposition fluxes in both the cities examined in this work are dominated by the particle phase. The values of the wet deposition fluxes in both areas are also similar to that in the ambient air of southern Taiwan (3.89–265 pg WHO_{2005}-TEQ m^{-2} $month^{-1}$ during 2012, 0.152–211 pg WHO_{2005}-TEQ m^{-2} $month^{-1}$ during 2013) (Tseng *et al*., 2014). The amount of precipitation in Nanjing was less than that in Guangzhou, but the amount of wet deposition was similar, thus demonstrating that wet deposition is not only affected by rainfall, but also by factors such as particulate matter concentration, temperature, and wind speed (Wang *et al*., 2010; Huang *et al*., 2011b).

The scavenging ratios of total-PCDD/Fs-WHO_{2005}-TEQ are presented in Fig. 5. The total scavenging ratios (S_{tot}) increase with temperature in both cities (Bidleman, 1988; Mandalakis and Stephanou, 2004; Tseng *et al*., 2014). In Guangzhou, the S_{tot} of total-PCDD/Fs-WHO_{2005}-TEQ ranged between 16350 to 34970, with an average of 24080. While in Nanjing, scavenging ratios (S_{tot}) of total-PCDD/Fs-WHO_{2005}-TEQ ranged between 19960 to 39990, with an average of 30950 during 2014. This is because the annual average temperature in Nanjing (16.4°C) is lower than that in Guangzhou (21.7°C), and the fraction of total-PCDD/Fs-WHO_{2005}-TEQ in the particle phase in Nanjing (75%) is higher than that in Guangzhou (59%) (Chandra Suryani *et al*., 2015). Compared with previous studies, the scavenging ratios of total-PCDD/Fs-WHO_{2005}-TEQ are similar to the typical ratio of semi-volatile organic compounds, which is between 10^4 and 10^5 (Eitzer and Hites, 1989; Tseng *et al*., 2014; Chandra Suryani *et al*., 2015).

Total (Dry + Wet) Deposition

The monthly total (wet + dry) deposition fluxes of total-PCDD/Fs-WHO_{2005}-TEQ are shown in Fig. 6. The monthly total deposition fluxes of PCDD/Fs ranged from 97.7 (June) and 559 (January) pg total-PCDD/Fs-WHO_{2005}-TEQ m^{-2} $month^{-1}$, with an average of 253.6 pg total-PCDD/Fs-WHO_{2005}-TEQ m^{-2} $month^{-1}$ in Guangzhou. As for the seasonal variation, the total deposition fluxes of total-PCDD/Fs-WHO_{2005}-TEQ in spring, summer, autumn and winter are 314, 97.1, 188 and 415 pg total-PCDD/Fs-WHO_{2005}-TEQ m^{-2} $month^{-1}$, respectively. While in Nanjing, the monthly total deposition fluxes of total-PCDD/Fs-

Table 2. Comparison of PCDD/Fs-TEQ Wet Deposition Fluxes between Previous Studies and This Study.

Area	Location	Period	Wet Deposition	Reference
Taiwan	Industrial	2014	0–183 pg WHO_{2005}-TEQ m^{-2} $month^{-1}$	Lee *et al.*, 2016
Taiwan	Industrial	2015	0.5–140 pg WHO_{2005}-TEQ m^{-2} $month^{-1}$	Lee *et al.*, 2016
Taiwan	Rural	2009–2010	5.4–92.2 pg I-TEQ m^{-2} $month^{-1}$	Huang *et al.*, 2011b
Taiwan	Rural	2009–2010	5.5–120 pg I-TEQ m^{-2} $month^{-1}$	Huang *et al.*, 2011b
Taiwan	Coastal	2013	0.8–30.1 pg WHO_{2005}-TEQ m^{-2} $month^{-1}$	Chandra Suryani *et al.*, 2015
Taiwan	Mountain	2013	0.2–21.5 pg WHO_{2005}-TEQ m^{-2} $month^{-1}$	Chandra Suryani *et al.*, 2015
Taiwan	Rural	2014	0–69.7 pg WHO_{2005}-TEQ m^{-2} $month^{-1}$	Lee *et al.*, 2016
Taiwan	Rural	2015	1.8–71.3 pg WHO_{2005}-TEQ m^{-2} $month^{-1}$	Lee *et al.*, 2016
China	Guangzhou	2014	0.58–229 pg WHO_{2005}-TEQ m^{-2} $month^{-1}$	this study
China	Nanjing	2014	10.7–266 pg WHO_{2005}-TEQ m^{-2} $month^{-1}$	this study
Taiwan	Rural	2012	3.89–265 pg WHO_{2005}-TEQ m^{-2} $month^{-1}$	Tseng *et al.*, 2014
Taiwan	Rural	2013	0.152–211 pg WHO_{2005}-TEQ m^{-2} $month^{-1}$	Tseng *et al.*, 2014

Table 3. Comparison of PCDD/Fs-TEQ Concentration in the Rain between Previous Studies and This Study.

Country	Location	Sampling Period	Concentration in the Rain	Reference
Taiwan	Kaohsiung	2014	0.263 pg WHO_{2005}-TEQ L^{-1}	Lee *et al.*, 2016
Taiwan	Kaohsiung	2015	0.307 pg WHO_{2005}-TEQ L^{-1}	Lee *et al.*, 2016
Taiwan	Meinong	2014	0.166 pg WHO_{2005}-TEQ L^{-1}	Lee *et al.*, 2016
Taiwan	Meinong	2015	0.167 pg WHO_{2005}-TEQ L^{-1}	Lee *et al.*, 2016
Taiwan	Xiaogang	2014	0.308 pg WHO_{2005}-TEQ L^{-1}	Lee *et al.*, 2016
Taiwan	Xiaogang	2015	0.369 pg WHO_{2005}-TEQ L^{-1}	Lee *et al.*, 2016
Taiwan	Hengchun	2012–2013	0.064 pg WHO_{2005}-TEQ L^{-1}	Chandra Suryani *et al.*, 2015
Taiwan	Lulin	2012–2013	0.027 pg WHO_{2005}-TEQ L^{-1}	Chandra Suryani *et al.*, 2015
China	Guangzhou	2014	0.49 pg WHO_{2005}-TEQ L^{-1}	this study
China	Nanjing	2014	1.7 pg WHO_{2005}-TEQ L^{-1}	this study

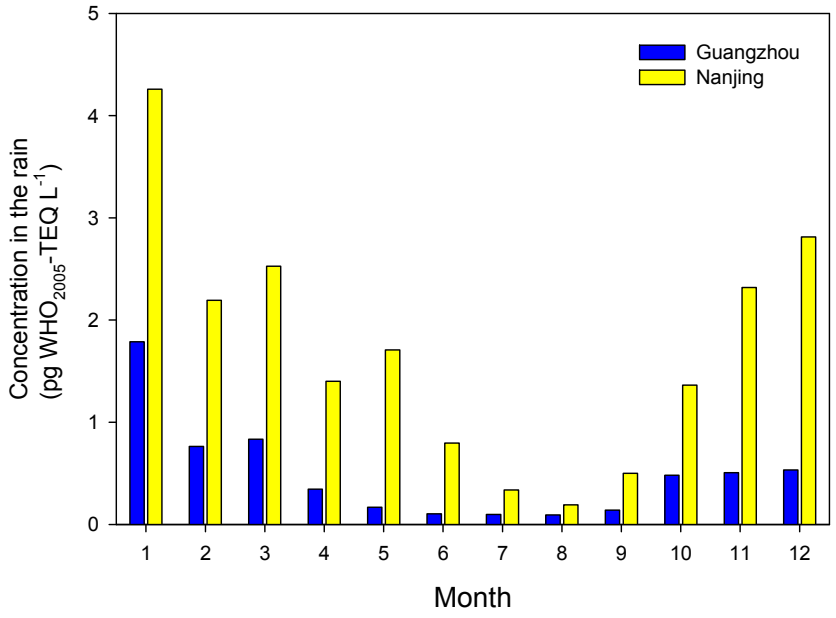

Fig. 4. Monthly Average Concentration of total-PCDD/Fs-WHO_{2005}-TEQ in the Rain.

WHO_{2005}-TEQ ranged from 135 (August) and 1250 (January) pg total-PCDD/Fs-WHO_{2005}-TEQ m^{-2} $month^{-1}$, with an average of 643 pg total-PCDD/Fs-WHO_{2005}-TEQ m^{-2} $month^{-1}$, respectively. As for the seasonal variation, the total deposition fluxes of total-PCDD/Fs-WHO_{2005}-TEQ in spring, summer, autumn and winter were 744, 277, 582 and 970 pg WHO_{2005}-TEQ m^{-2} $month^{-1}$, respectively. Comparisons between the total (dry + wet) depositions simulated in this study and in other works are shown in Table 4.

Figs. 7(A) and 7(B) demonstrate the fractions of total-PCDD/Fs-WHO_{2005}-TEQ contributed by dry and wet deposition fluxes during 2014 in Guangzhou and Nanjing,

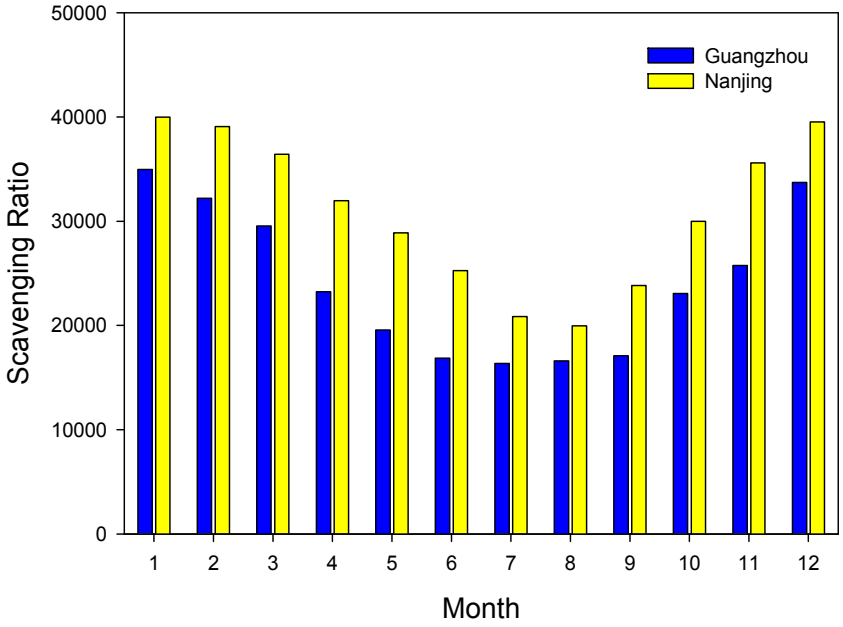

Fig. 5. Monthly Average Scavenging Ratio of total-PCDD/Fs-WHO_{2005}-TEQ during 2014.

Table 4. Comparison of Total (Dry + Wet) PCDD/Fs-TEQ Deposition Fluxes between Previous Studies and This Study.

Area	Location	Sampling Period	Total (Dry + Wet) Deposition (TEQ)	Reference
Taiwan	Rural	2009–2010	5020–5110 pg I-TEQ m^{-2} $year^{-1}$	Huang *et al.*, 2011b
Taiwan	Coastal	2013	657 pg WHO_{2005}-TEQ m^{-2} $year^{-1}$	Chandra Suryani *et al.*, 2015
Taiwan	Mountain	2013	249 pg WHO_{2005}-TEQ m^{-2} $year^{-1}$	Chandra Suryani *et al.*, 2015
Tokyo	Urban	2001	17000 pg I-TEQ m^{-2} $year^{-1}$	Ogura *et al.*, 2001
Yokohama	Urban	2001	11000 pg I-TEQ m^{-2} $year^{-1}$	Ogura *et al.*, 2001
Tanzawa	Mountain	2001	5700 pg I-TEQ m^{-2} $year^{-1}$	Ogura *et al.*, 2001
France	Lagoon	2011	4280 pg I-TEQ m^{-2} $year^{-1}$	Guerzoni *et al.*, 2004
Italy	Lake	2005	140 pg WHO_{2005}-TEQ m^{-2} $week^{-1}$	Castro-Jiménez *et al.*, 2008
China	Guangzhou	2014	3043 pg WHO_{2005}-TEQ m^{-2} $year^{-1}$	this study
China	Nanjing	2014	7610 pg WHO_{2005}-TEQ m^{-2} $year^{-1}$	this study

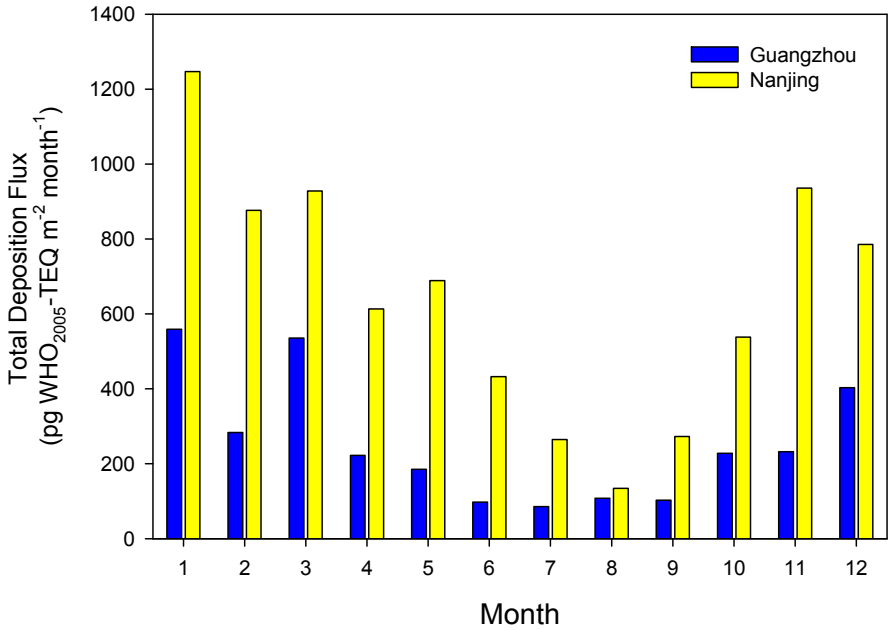

Fig. 6. Monthly Average Total (Dry + Wet) Deposition Flux of total-PCDD/Fs-WHO_{2005}-TEQ during 2014.

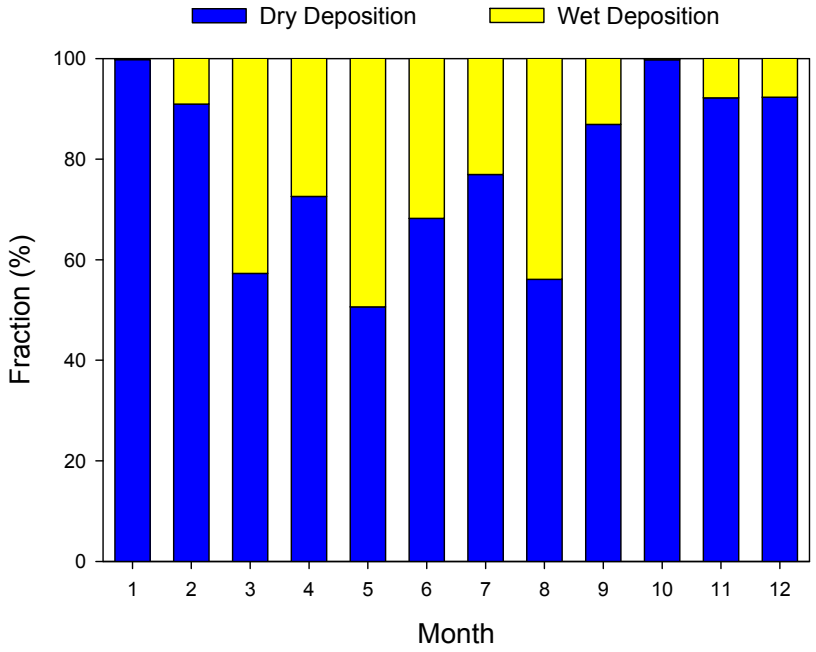

Fig. 7(A). The Fraction of Total Deposition Flux in total-PCDD/Fs-WHO_{2005}-TEQ Contributed by the Dry and Wet Deposition, respectively, in Guangzhou.

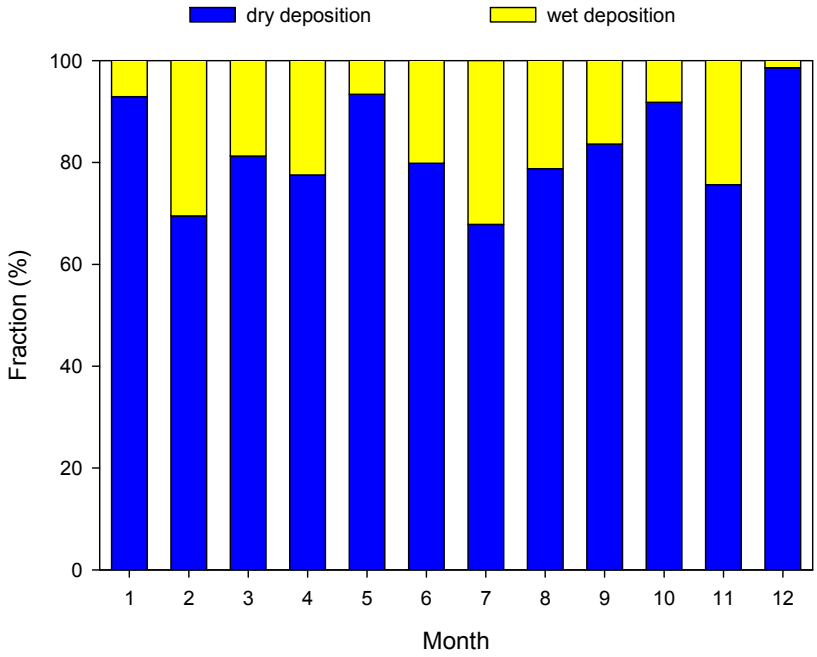

Fig. 7(B). The Fraction of Total Deposition Flux in total-PCDD/Fs-WHO_{2005}-TEQ Contributed by the Dry and Wet Deposition, respectively, in Nanjing.

between 50.6% (May) and 99.8% (January), with an average of 78.6% in Guangzhou. However, in Nanjing, the fraction of dry deposition fluxes ranged between 66.5% (July) and 98.6% (December), with an average of 82.4%. As the results show, the higher fraction contributed by dry deposition usually occurs in months with less rainfall, and vice versa. This suggests that the intensified rainfall may cause the low fraction of total-PCDD/Fs-WHO_{2005}-TEQ contributed by dry deposition. The results also demonstrate that the contributions of dry deposition fluxes are much greater than those of wet deposition fluxes in both cities. The dry deposition fluxes are the major removal mechanism for the total-PCDD/Fs-WHO_{2005}-TEQ in the ambient air. Therefore, the control of $PM_{2.5}$ emissions is of great importance in reducing the atmospheric deposition of total-PCDD/Fs-WHO_{2005}-TEQ. The relevant previous studies published in *Aerosol and Air Quality Research,* which are cited in this study are listed in Table 5 for reference.

Table 5. The Relevant Previous Studies Published in *Aerosol and Air Quality Research*.

Reference	Object	Area
Lee *et al.*, 2003	Emission	Taiwan
Kao *et al.*,2006	Emission	Taiwan
Hu *et al.*, 2009	Emission	Taiwan
Wang *et al.*, 2010	Dry Deposition and Wet Deposition	Taiwan
Huang *et al.*, 2011a	Dry deposition	Taiwan
Huang *et al.*, 2011b	Wet deposition	Taiwan
Mi *et al.*, 2012	Dry deposition	Taiwan
Yu *et al.*, 2013	Emission	China
Guo *et al.*, 2014	Emission	China
Lin *et al.*, 2014	Emission	China
Chandra Suryani *et al.*, 2015	Dry deposition and Wet deposition	Taiwan
Chow *et al.*, 2015	Concentration	America
Cheruiyot *et al.*, 2015	Emission	Taiwan
Zhang *et al.*, 2015	Concentration	China
Cheruiyot *et al.*, 2016	Emission	Taiwan
Chi *et al.*, 2016	Concentration	Vietnam/Taiwan
Lee *et al.*, 2016	Dry Deposition and Wet Deposition	Taiwan
Lu *et al.*, 2016	Emission	Taiwan
Tang *et al.*, 2017	Concentration	China

CONCLUSION

This study investigated the atmospheric deposition of PCDD/Fs during 2014 in Nanjing and Guangzhou, and the results of this study can be summarized as follows:

1. During 2014, the monthly dry deposition fluxes of total-PCDD/Fs-WHO_{2005}-TEQ ranged between 60.6–558 and 104–1160 pg total-PCDD/Fs-WHO_{2005}-TEQ m^{-2} $month^{-1}$ in Guangzhou and Nanjing, respectively. Dry deposition fluxes in Nanjing (averaged 540 pg WHO_{2005}-TEQ m^{-2} $month^{-1}$) were approximately 2.6 times of magnitude higher than those in Guangzhou (206 pg WHO_{2005}-TEQ m^{-2} $month^{-1}$).
2. The annual dry deposition fluxes of total-PCDD/Fs-WHO_{2005}-TEQ were 2470 and 6480 pg WHO_{2005}-TEQ m^{-2} $year^{-1}$ in Guangzhou and Nanjing, respectively. A lower average temperature and higher $PM_{2.5}$ concentration resulted in a higher dry deposition flux of total-PCDD/Fs-WHO_{2005}-TEQ in Nanjing. The total dry deposition fluxes of total-PCDD/Fs-WHO_{2005}-TEQ depended significantly on the particle phase partition in the atmosphere. The higher the fraction of the particle phase, the greater the total dry deposition fluxes of the total-PCDD/Fs-WHO_{2005}-TEQ.
3. The dry deposition velocities of particle phase total-PCDD/Fs-WHO_{2005}-TEQ in Guangzhou (0.49–0.98 cm s^{-1}) and Nanjing (0.44–0.8 cm s^{-1}) were similar to those found in ambient air near MSWIs in Taiwan.
4. The wet deposition fluxes of total-PCDD/Fs-WHO_{2005}-TEQ were in the range of 0.58–229 and 10.7–266 pg WHO_{2005}-TEQ m^{-2} $month^{-1}$ in Guangzhou and Nanjing, respectively.
5. Monthly concentrations of total-PCDD/Fs-WHO_{2005}-TEQ in rain were in the range of 0.092–1.79 and 0.191–4.26 pg WHO_{2005}-TEQ L^{-1} in Guangzhou and Nanjing, respectively.
6. The total scavenging ratios of total-PCDD/Fs-WHO_{2005}-TEQ increased with an increase in temperature in both cities, and the scavenging ratio in Guangzhou (average 30950) was higher than that in Nanjing (average 24080). This is because Nanjing City has a lower annual average temperature and a higher fraction of PCDD/Fs-WHO_{2005}-TEQ in the particle phase than Guangzhou.
7. The total (dry + wet) deposition fluxes of total-PCDD/Fs-WHO_{2005}-TEQ ranged from 97.7 to 559 and from 135 to 1250 pg WHO_{2005}-TEQ m^{-2} $month^{-1}$ in Guangzhou and Nanjing, respectively.
8. The fraction total (dry + wet) deposition fluxes of total-PCDD/Fs-WHO_{2005}-TEQ contributed by dry deposition were 78.6% and 82.4% in Guangzhou and Nanjing, respectively. Dry deposition flux is thus the major mechanism for removal of PCDD/Fs from the atmosphere in both cities.

ACKNOWLEDGMENTS

This research was supported by the Fundamental Research Funds for the Central Universities.

REFERENCES

Addink, R. and Altwicker, E.R. (2001). Formation of polychlorinated dibenzo-*p*-dioxins/dibenzofurans from residual carbon on municipal solid waste incinerator fly ash using $Na^{37}Cl$. *Chemosphere* 44: 1361–1367.

Bidleman, T.F. (1988). Atmospheric processes. *Environ. Sci. Technol.* 22: 361–367.

Bumb, R., Crummett, W., Cutie, S., Gledhill, J., Hummel, R., Kagel, R. and Lamparski, L. (1980). Trace chemistries of fire: A source of chlorinated dioxins. *Science* 210: 385–390.

Castro-Jiménez, J., Mariani, G., Eisenreich, S., Christoph,

E., Hanke, G., Canuti, E., Skejo, H. and Umlauf, G. (2008). Atmospheric input of POPs into Lake Maggiore (Northern Italy): PCDD/F and dioxin-like PCB profiles and fluxes in the atmosphere and aquatic system. *Chemosphere* 73: S122–S130.

Chandra Suryani, R., Lee, W.J., Endah Mutiara, M., Mwangi, J.K., Wang, L.C., Lin, N.H. and Chang-Chien, G.P. (2015). Atmospheric deposition of polychlorinated dibenzo-*p*-dioxins and dibenzofurans at coastal and high mountain areas in Taiwan. *Aerosol Air Qual. Res.* 15: 1390–1411.

Chang, M.O., Chow, J.C., Watson, J.G., Hopke, P.K., Yi, S.M. and England, G.C. (2004). Measurement of ultrafine particle size distributions from coal-, oil-, and gas-fired stationary combustion sources. *J. Air Waste Manage. Assoc.* 54: 1494–1505.

Cheruiyot, N.K., Lee, W.J., Mwangi, J.K., Wang, L.C., Lin, N.H., Lin, Y.C., Cao, J., Zhang, R. and Chang-Chien, G.P. (2015). An overview: Polycyclic aromatic hydrocarbon emissions from the stationary and mobile sources and in the ambient air. *Aerosol Air Qual. Res.* 15: 2730–2762.

Cheruiyot, N.K., Lee, W.J., Yan, P., Mwangi, J.K., Wang, L.C., Gao, X., Lin, N.H. and Chang-Chien, G.P. (2016). An overview of PCDD/F inventories and emission factors from stationary and mobile sources: What we know and what is missing. *Aerosol Air Qual. Res.* 16: 2965–2988.

Chi, K.H., Hung, N.T., Lin, C.Y., Wang, S.H., Ou-Yang, C.F., Lee, C.T. and Lin, N.H. (2016). Evaluation of atmospheric PCDD/Fs at two high-altitude stations in Vietnam and Taiwan during Southeast Asia biomass burning. *Aerosol Air Qual. Res.* 16: 2706–2715.

Chow, J.C., Yang, X., Wang, X., Kohl, S.D., Hurbain, P.R., Chen, L.A. and Watson, J.G. (2015). Characterization of ambient PM_{10} bioaerosols in a California agricultural town. *Aerosol Air Qual. Res.* 15: 1433–1447.

Correa, O., Raun, L., Rifai, H., Suarez, M., Holsen, T. and Koenig, L. (2006). Depositional flux of polychlorinated dibenzo-*p*-dioxins and polychlorinated dibenzofurans in an urban setting. *Chemosphere* 64: 1550–1561.

Eitzer, B.D. and Hites, R.A. (1989). Atmospheric transport and deposition of polychlorinated dibenzo-*p*-dioxins and dibenzofurans. *Environ. Sci. Technol.* 23:1396–1401.

Gao, H., Ni, Y., Zhang, H., Zhao, L., Zhang, N., Zhang, X., Zhang, Q. and Chen, J. (2009). Stack gas emissions of PCDD/Fs from hospital waste incinerators in China. *Chemosphere* 77: 634–639.

Guerzoni, S., Rossini, P., Molinaroli, E., Rampazzo, G. and Raccanelli, S. (2004). Measurement of atmospheric deposition of polychlorinated dibenzo-*p*-dioxins and dibenzofurans in the Lagoon of Venice, Italy. *Chemosphere* 54: 1309–1317.

Guo, C.J., Wang, M.S., Lin, S.L., Mi, H.H., Wang, L.C. and Chang-Chien, G.P. (2014). Emissions of PCDD/Fs and PCBs during the cold start-up of municipal solid waste incinerators. *Aerosol Air Qual. Res.* 14: 1593–1604.

Hu, M.T., Chen, S.J., Lai, Y.C., Huang, K.L., Chang-Chien, G.P. and Tsai, J.H. (2009). Characteristics of polychlorinated dibenzo-*p*-dioxins/dibenzofuran from joss paper burned in Taiwanese temples. *Aerosol Air Qual. Res.* 9: 369–377.

Huang, C.J., Chen, K.S., Lai, Y.C., Wang, L.C. and Chang-Chien, G.P. (2011a). Characterization of atmospheric dry deposition of polychlorinated dibenzop-dioxins/dibenzofuran in a rural area of taiwan. *Aerosol Air Qual. Res.* 11: 448–459.

Huang, C.J., Chen, K.S., Lai, Y.C., Wang, L.C. and Chang-Chien, G.P. (2011b). Wet deposition of polychlorinated dibenzo-*p*-dioxins/dibenzofuran in a rural area of Taiwan. *Aerosol Air Qual. Res.* 11: 732–748.

Kao, J.H., Chen, K.S., Chang-Chien, G.P. and Chou, I.C. (2006). Emissions of polychlorinated dibenzo-*p*-dioxins and dibenzofurans from various stationary sources. *Aerosol Air Qual. Res.* 6: 170–179.

Kim, E.J., Oh, J.E. and Chang, Y.S. (2003). Effects of forest fire on the level and distribution of PCDD/Fs and PAHs in soil. *Sci. Total Environ.* 311: 177–189.

Koester, C.J. and Hites, R.A. (1992). Wet and dry deposition of chlorinated dioxins and furans. *Environ. Sci. Technol.* 26: 1375–1382.

Laden, F., Neas, L.M., Dockery, D.W. and Schwartz, J. (2000). Association of fine particulate matter from different sources with daily mortality in six US cities. *Environ. Health Perspect.* 108: 941–947.

Lee, K.L., Lee, W.J., Mwangi, J.K., Wang, L.C., Gao, X. and Chang-Chien, G.P. (2016). Atmospheric $PM_{2.5}$ and depositions of polychlorinated dibenzo-*p*-dioxins and dibenzofurans in Kaohsiung area, southern Taiwan. *Aerosol Air Qual. Res.* 16: 1775–1791.

Lee, S.J., Ale, D., Chang, Y.S., Oh, J.E. and Shin, S.K. (2008). Seasonal and particle size-dependent variations in gas/particle partitioning of PCDD/Fs. *Environ. Pollut.* 153: 215–222.

Lee, W.J., Lewis, S.J.L., Chen, Y.Y., Wang, Y.F., Sheu, H.L., Su, C.C. and Fan, Y.C. (1996). Polychlorinated biphenyls in the ambient air of petroleum refinery, urban and rural areas. *Atmos. Environ.* 30: 2371–2378.

Lee, W.J., Shih, S.I., Li, H.W., Lin, L.F., Yu, K.M., Lu, K., Wang, L.C., Chang-Chien, G.P., Fang, K. and Lin, M. (2009). Assessment of polychlorinated dibenzo-*p*-dioxins and dibenzofurans contribution from different media to surrounding duck farms. *J. Hazard. Mater.* 163: 1185–1193.

Lee, W.S., Chang-Chien, G.P., Wang, L.C., Lee, W.J., Tsai, P.J. and Chen, C.K. (2003). Emissions of polychlorinated dibenzo-*p*-dioxins and dibenzofurans from the incinerations of both medical and municipal solid wastes. *Aerosol Air Qual. Res.* 3: 1–6.

Ligocki, M.P., Leuenberger, C. and Pankow, J.F. (1985a). Trace organic compounds in rain—II. Gas scavenging of neutral organic compounds. *Atmos. Environ.* 19: 1609–1617.

Ligocki, M.P., Leuenberger, C. and Pankow, J.F. (1985b). Trace organic compounds in rain—III. Particle scavenging of neutral organic compounds. *Atmos. Environ.* 19: 1619–1626.

Lin, X., Huang, Q., Chen, T., Li, X., Lu, S., Wu, H., Yan, J., Zhou, M. and Wang, H. (2014). PCDD/F and PCBz

emissions during start-up and normal operation of a hazardous waste incinerator in China. *Aerosol Air Qual. Res*. 14: 1142–1151.

Liu, J., Xu, X., Gao, L., Zhong, G., He, C., Ye, W., Ma, Z. and Pang, X. (2006). Assessment of dioxin pollution in part of medical solid waste incinerator and analysis of the dioxin emission quantity of China. Persistent Organic Pollutants Forum 2006 and the 1st National Symposium on Persistent Organic Pollutants, Beijing, China, pp. 107–110.

Lohmann, R. and Jones, K.C. (1998). Dioxins and furans in air and deposition: A review of levels, behaviour and processes. *Sci. Total Environ.*219: 53–81.

Lohmann, R., Green, N.J. and Jones, K.C. (1999). Detailed studies of the factors controlling atmospheric PCDD/F concentrations. *Environ. Sci. Technol*. 33: 4440–4447.

Lu, H.Y., Lin, S.L., Mwangi, J.K., Wang, L.C. and Lin, H.Y. (2016). Characteristics and source apportionment of atmospheric $PM_{2.5}$ at a coastal city in southern Taiwan. *Aerosol Air Qual. Res*. 16: 1022–1034.

Mandalakis, M. and Stephanou, E.G. (2004). Wet deposition of polychlorinated biphenyls in the eastern Mediterranean. *Environ. Sci. Technol*. 38: 3011–3018.

Meng, B., Ma, W.L., Liu, L.Y., Zhu, N.Z., Song, W.W., Lo, C.Y., Li, J., Kannan, K. and Li, Y.F. (2016). PCDD/Fs in soil and air and their possible sources in the vicinity of municipal solid waste incinerators in northeastern China. *Atmos. Pollut. Res.* 7: 355–362.

Mi, H.H., Wu, Z.S., Lin, L.F., Lai, Y.C., Lee, Y.Y., Wang, L.C. and Chang-Chien, G.P. (2012). Atmospheric dry deposition of polychlorinated dibenzo-*p*-dioxins/dibenzofurans (PCDD/Fs) and polychlorinated biphenyls (PCBs) in southern Taiwan. *Aerosol Air Qual. Res*. 12: 1016–1029.

Moon, H.B., Lee, S.J., Choi, H.G. and Ok, G. (2005). Atmospheric deposition of polychlorinated dibenzo-*p*-dioxins (PCDDs) and dibenzofurans (PCDFs) in urbanand suburban areas of Korea. *Chemosphere* 58: 1525–1534.

Neuer-Etscheidt, K., Nordsieck, H.O., Liu, Y., Kettrup, A. and Zimmermann, R. (2006). PCDD/F and other micropollutants in MSWI crude gas and ashes during plant start-up and shut-down processes. *Environ. Sci. Technol*. 40: 342–349.

Ogura, I., Masunaga, S. and Nakanishi, J. (2001). Atmospheric deposition of polychlorinated dibenzo-*p*-dioxins, polychlorinated dibenzofurans, and dioxin-like polychlorinated biphenyls in the Kanto Region, Japan. *Chemosphere* 44: 1473–1487.

Oh, J.E., Choi, J.S. and Chang, Y.S. (2001). Gas/particle partitioning of polychlorinated dibenzo-*p*-dioxins and dibenzofurans in atmosphere; evaluation of predicting models. *Atmos. Environ*. 35: 4125–4134.

Oh, J.E., Lee, K.T., Lee, J.W. and Chang, Y.S. (1999). The evaluation of PCDD/Fs from various Korean incinerators. *Chemosphere* 38: 2097–2108.

Prange, J.A., Gaus, C., Päpke, O. and Müller, J.F. (2002). Investigations into the PCDD contamination of topsoil, river sediments and kaolinite clay in Queensland, Australia. *Chemosphere* 46: 1335–1342.

Prange, J.A., Gaus, C., Weber, R., Päpke, O. and Müller, J.F. (2003). Assessing forest fire as a potential PCDD/F source in Queensland, Australia. *Environ. Sci. Technol*. 37: 4325–4329.

Sheu, H.L., Lee, W.J., Su, C.C., Chao, H.R. and Fan, Y.C. (1996). Dry deposition of polycyclic aromatic hydrocarbons in ambient air. *J. Environ. Eng.* 122: 1101–1109.

Shih, M., Lee, W.S., Chang Chien, G.P., Wang, L.C., Hung, C.Y. and Lin, K.C. (2006). Dry deposition of polychlorinated dibenzo-*p*-dioxins and dibenzofurans (PCDD/Fs) in ambient air. *Chemosphere* 62: 411–416.

Sugita, K., Asada, S., Yokochi, T., Okazawa, T., Ono, M. and Goto, S. (1994). Survey of polychlorinated dibenzo-*p*-dioxins, polychlorinated dibenzofurans and polychlorinated biphenyls in urban air. *Chemosphere* 29: 2215–2221.

Tang, H., Cui, K., Xing, J., Zhu, J., Lee, W.J., Mwangi, J.K. and Lee, Y.C. (2017). Part I: $PM_{2.5}$ and polychlorinated dibenzo-*p*-dioxins and dibenzofurans(PCDD/Fs) in the ambient air of Southern China. *Aerosol Air Qual. Res*. 17: 1450–1469.

Tian, H. and Ouyang, N. (2003). Preliminary investigation on dioxin emissions from MSW incinerators. *Huanjing Huaxue* 22: 255–258.

Tseng, Y.J., Mi, H.H., Hsieh, L.T., Liao, W.T. and Chang-Chien, G.P. (2014). Atmospheric deposition modeling of polychlorinated dibenzo-*p*-dioxins, dibenzofurans and polychlorinated biphenyls in the ambient air of southern Taiwan. Part II. Wet depositions and total deposition fluxes. *Aerosol Air Qual. Res*. 14: 1966–1985.

Wang, G., Cheng, S., Li, J., Lang, J., Wen, W., Yang, X. and Tian, L. (2015). Source apportionment and seasonal variation of $PM_{2.5}$ carbonaceous aerosol in the Beijing-Tianjin-Hebei Region of China. *Environ. Monit. Assess*. 187: 143.

Wang, Q., Jin, Y., Li, X., Chen, J., Lu, S., Chen, T., Yan, J., Zhou, M. and Wang, H. (2014). PCDD/F emissions from hazardous waste incinerators in China. *Aerosol Air Qual. Res*. 14: 1152–1159.

Wang, Y.F., Hou, H.C., Li, H.W., Lin, L.F., Wang, L.C., Chang-Chien, G.P. and You, Y.S. (2010). Dry and wet depositions of polychlorinated dibenzo-*p*-dioxins and dibenzofurans in the atmosphere in Taiwan. *Aerosol Air Qual. Res*. 10: 378–390.

Wu, Y.L., Lin, L.F., Hsieh, L.T., Wang, L.C. and Chang-Chien, G.P. (2009). Atmospheric dry deposition of polychlorinated dibenzo-*p*-dioxins and dibenzofurans in the vicinity of municipal solid waste incinerators. *J. Hazard. Mater*. 162: 521–529.

Xu, P., Chen, Y. and Ye, X. (2013). Haze, air pollution, and health in China. *The Lancet* 382: 2067.

Yu, L., Wang, G., Zhang, R., Zhang, L., Song, Y., Wu, B., Li, X., An, K. and Chu, J. (2013). Characterization and source apportionment of $PM_{2.5}$ in an urban environment in Beijing. *Aerosol Air Qual. Res*. 13: 574–583.

Zhang X, Zhu Q.Q., Dong S.J., Zhang H.X., Wang X.K., Wang M., Gao L.R. and Zheng M.H. (2015). Particle size distributions of PCDD/Fs and PBDD/Fs in ambient air in a suburban area in Beijing, China. *Aerosol Air Qual. Res*. 15: 1933–1943.

Zhang, B., Meng, F., Shi, C., Yang, F., Wen, D., Aronsson, J., Gbor, P.K. and Sloan, J.J. (2009). Modeling the atmospheric transport and deposition of polychlorinated dibenzo-*p*-dioxins and dibenzofurans in North America. *Atmos. Environ*. 43: 2204–2212.

Received for review, May 21, 2017
Revised, June 19, 2017
Accepted, June 20, 2017