Occurrence, phase distribution and depositional intensity of dichlorodiphenyltrichloroethane (DDT) and its metabolites in air and precipitation of the Pearl River Delta, China

Qiang Yue [a,b], Kai Zhang [a,d], Bao-Zhong Zhang [a], Shao-Meng Li [c], Eddy Y. Zeng [a,*]

[a] State Key Laboratory of Organic Geochemistry, Guangzhou Institute of Geochemistry, Chinese Academy of Sciences, Guangzhou 510640, China
[b] Yingdong College of Life Science, Shaoguan University, Shaoguan, 512005, China
[c] Environment Canada, 4905 Dufferin Street, Toronto, Ontario M3H 5T4, Canada
[d] Graduate School of Chinese Academy of Sciences, Beijing 100049, China

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ABSTRACT

Concentrations of dichlorodiphenyltrichloroethane (DDT) and its metabolites (designated as DDTs, the sum of o,p'- and p,p'-DDT, o,p'- and p,p'-DDE, and o,p'- and p,p'-DDD) in air and precipitation from the Pearl River Delta (PRD) of China were determined. Total concentrations of DDTs in air (gas + particle) and precipitation (dissolved + particle) were 170 ± 120 pg m^{-3} and 940 ± 180 pg L^{-1} for Dongguan (rural) and 240 ± 120 pg m^{-3} and 790 ± 140 pg L^{-1} for Shunde (rural), respectively, while they were 1550 ± 640 pg L^{-1} in precipitation from Guangzhou (urban). Log-transformed partition coefficients between air and particulate organic matter (log K'_{oa}) of p,p'-DDT, p,p'-DDE, p,p'-DDD, o,p'-DDT, o,p'-DDE, and o,p'-DDD were 9.64 ± 0.58, 10.07 ± 0.56, 9.90 ± 0.76, 10.06 ± 0.66, 10.02 ± 0.72, and 10.13 ± 0.57, respectively; while those between water and particulate organic matter (log K'_{om}) were 6.58 ± 0.66, 6.36 ± 0.53, 6.01 ± 0.62, 6.41 ± 0.42, 5.98 ± 0.76, and 5.95 ± 0.66, respectively. Total washout ratios by bulk rainfalls ranged from 4600 for o,p'-DDT to 54,000 for p,p'-DDT. Estimated average dry particle and wet depositional intensities in the PRD were 2.1×10^{-6} and 1.6×10^{-6} g m^{-2} y^{-1}.

1. Introduction

Protection of the environment from contamination by persistent organic pollutants (POPs) has been a central task for the global community under the Stockholm Convention (Stockholm Convention, 2001). One of the characteristic properties of POPs is their potential for long-range atmospheric transport that has been demonstrated to spread POPs globally (Wania and Mackay, 1996). At the same time, atmospheric POPs can also be removed via various routes, such as dry particle deposition and wet deposition via rain (Venier and Hites, 2008; Wania and Westgate, 2008; Zhang et al., 2009). On a regional scale, the occurrence and transport efficiency of POPs in the atmosphere dictate the global fate of POPs, and are influenced by removal mechanisms and meteorological conditions.

Dichlorodiphenyltrichloroethane (DDT) was phased out as a pesticide in the mid 1980s on a global scale (Guo et al., 2009). However, residues of DDT and its metabolites (designated as DDTs, sum of o,p'- and p,p'-DDT, o,p'- and p,p'-DDE, and o,p'- and p,p'-DDD) have been detected in the global environment (Guglielmo et al., 2009; van den Brink et al., 2009). Although tremendous efforts have been directed toward monitoring atmospheric deposition of DDTs in the Great Lakes areas (Buehler et al., 2003) and New Jersey (Reinfelder et al., 2004) since the 1990s, limited work on this topic has been done in China.

The Pearl River Delta (PRD), a typical subtropical watershed in South China (Fig. 1), has long been a highly developed agricultural zone. Large amounts of previously applied DDT in the PRD have resulted in the occurrence of abundant DDTs in various environmental compartments (Guo et al., 2009), particularly in the atmosphere (Wang et al., 2007; Choi et al., 2009; Liu et al., 2009). Yet, aerial deposition of DDTs in the PRD has not been adequately investigated and only a few reports are available in the literature. Wong et al. (2004) obtained atmospheric depositional rates of DDTs in Hong Kong from 2002 to 2003, while Li et al. (2010) measured the bulk particle depositional rates of DDTs in the PRD in 2002. On the other hand, no reports of the occurrence of DDTs in precipitation and dry or wet depositional rates of DDTs in the PRD have been published. To fill the knowledge gap in understanding the environmental fate of DDTs, the present study was aimed to estimate washout ratios and atmospheric depositional intensities of DDTs in rural (Dongguan and Shunde; Fig. 1) and urban (Guangzhou; Fig. 1) areas, based on a year-long, seasonal sampling campaign.

* Corresponding author. Tel.: +86 20 85291421; fax: +86 20 85290706.
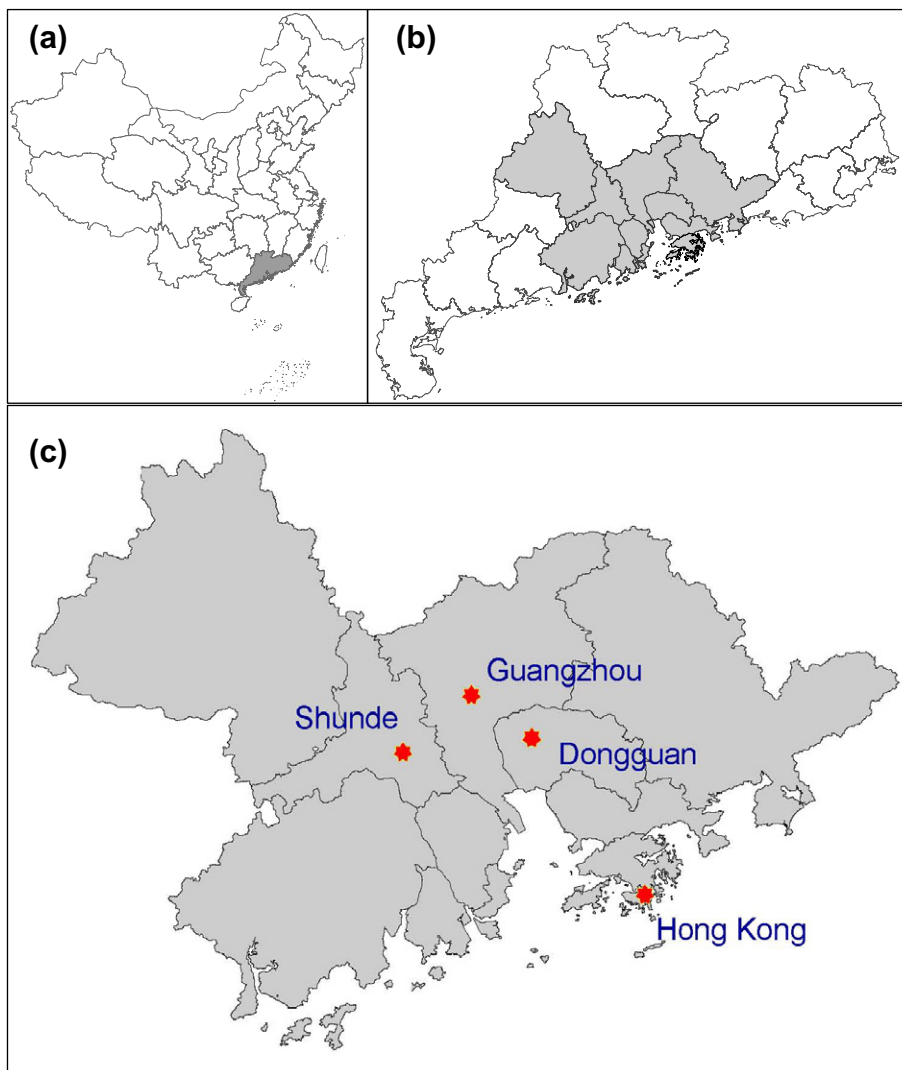E-mail address: eddyzeng@gig.ac.cn (E.Y. Zeng).

Fig. 1. Map showing (a) China; (b) the Pearl River Delta; and (c) locality of air (Shunde and Dongguan) and bulk precipitation (Shunde, Dongguan, and Guangzhou) sampling sites.

2. Materials and methods

2.1. Sampling procedures

A total of 34 air and 12 precipitation samples were collected at two rural sites in Dongguan and Shunde from October 2006 to September 2007 (Tables S1 and S2, Supplementary material). Eleven precipitation samples were collected at an urban site in Guangzhou during the same time period (Tables S2, Supplementary material). The sampling procedures described previously (Zhang et al., 2009) are briefly stated here. Clean glass fiber filter (GFF; 20.3 × 25.4 cm with 0.6-μm nominal pore size; Whatman International, Maidstone, England) and PUF (6.5-cm diameter and 8.0 cm thickness with a density of 0.030 g cm^{-3}) plug installed in high-volume air samplers were employed to collect suspended particle and gaseous samples, respectively, every 24 h. For sampling of precipitation, 20–90 L of rainwater collected with stainless steel containers were transferred to pre-cleaned brown glass bottles. All air and precipitation samples were cooled with ice as soon as they were collected and were immediately transported to the laboratory.

2.2. Sample extraction

Water samples were filtered with a peristaltic pump system, and suspended particulates were retained by another type of GFFs (142-mm diameter and 0.7-μm nominal pore size; Millipore, Billerica, MA, USA). Organic materials in filtrates were collected with a glass column (400 mm × 25 mm i.d.) packed with a mixture of XAD-2 and XAD-4 resin (1:1 in mass, ultrasonically cleaned with methanol prior to use). The resin was eluted three times each with 50 mL of methanol and then ultrasonically extracted with 50 mL of dichloromethane:methanol (1:1 in volume) mixture for three times. Extracts from the above steps were combined, and then spiked with known amounts of two surrogate standards (PCB 67 and PCB 191). The combined extract was diluted with 500 mL of distilled water, spiked with an appropriate amount of sodium chloride, and liquid–liquid extracted five times with 60 mL of DCM each. Half of the extract was concentrated with a Zymark TurboVap 500 (Zymark, Hopkinton, MA, USA), solvent-exchanged to hexane, and then concentrated to 1 mL. A glass column packed with alumina:silica gel (1:2 in volume) was used to fractionate the extract. The fraction containing DDTs was eluted with 70 mL of a mixture

of hexane: dichloromethane (1:1 in volume). This fraction was transferred to a special glass vial (~2 mL in volume), concentrated to 0.5 mL and spiked with the internal standard (PCB 82) prior to instrumental analysis.

Loaded GFF and PUF plugs (atmospheric gas- and particle-phase samples, respectively) were spiked with the surrogate standards prior to Soxhlet extraction with a mixture of acetone:hexane (1:1 in volume) for 48 h. Half of the extract was fractionated with the same method applied to the filtrate samples.

2.3. Instrumental analysis

Determination of DDTs was conducted with a Varian 3800 gas chromatograph interfaced with a Saturn 2000 mass spectrometer in the selective ion monitoring mode and equipped with a 60 m × 0.25 mm-i.d. (0.25 μm film thickness) DB-5MS column (J&W Scientific, Folsom, CA, USA). The detailed analytical and quantification procedures for DDTs were described previously (Meng et al., 2007).

2.4. Quality assurance and quality control

Field blanks (unloaded GFF and PUF brought to/from sampling sites were treated as field blanks for air samples; redistilled water brought to/from sampling sites was treated as field blanks for pre-cipitation samples), laboratory blanks (unloaded GFF and PUF and redistilled water) and spiked blanks (solvent spiked with a known amount of analytes) were analyzed for every nine field samples. The concentrations of the target analytes in blank samples were al-ways below the lowest concentration (10 ng mL^{-1}) of the calibra-tion curve. The recoveries of DDTs were 70–114% with relative standard deviations <15% in five spiked blanks. The recoveries of the surrogate standards in all samples were 91 ± 13% and 86 ± 19% for PCB 67 and PCB 191, respectively. All reported concen-trations were not corrected with either blank concentrations or recoveries of the surrogate standards.

2.5. Data analysis

All concentrations of DDTs in the particulate and dissolved phases of precipitation samples were adjusted with the corre-sponding volumes of rainfall to the volume weighted mean (VWM) concentrations. The VWM concentration and standard er-ror of mean (SEM) were calculated by the following equations (Endlich et al., 1988; Venier and Hites, 2008).

$$VWM = \Sigma C_i / \Sigma (C_i \cdot V_i) \tag{1}$$

SEM

$$= \left\{ \frac{N}{(N-1)(\sum V_i)^2} \left[\sum (V_i C_i - \bar{V}\bar{C})^2 - 2\bar{C}\sum (V_i - \bar{V})(V_i C_i - \bar{V}\bar{C}) + \bar{C}^2 \sum (V_i - \bar{V})^2 \right] \right\}^{1/2}$$

where C_i is the DDT concentration in sample i with the volume rep-resented by V_i, N is the total number of samples, and an average was calculated for each sampling site.

3. Results and discussion

3.1. Occurrence of dichlorodiphenyltrichloroethane and its metabolites in air and precipitation

Concentrations of DDTs in the particulate and gaseous phases of the air samples were 22 ± 23 (mean ± SD) pg m^{-3} and 180 ± 120 pg m^{-3}, respectively, indicating that DDTs were mainly distributed in the gaseous phase. Total concentrations of DDTs in the air (particulate plus gas phases) from Dongguan (170 ±
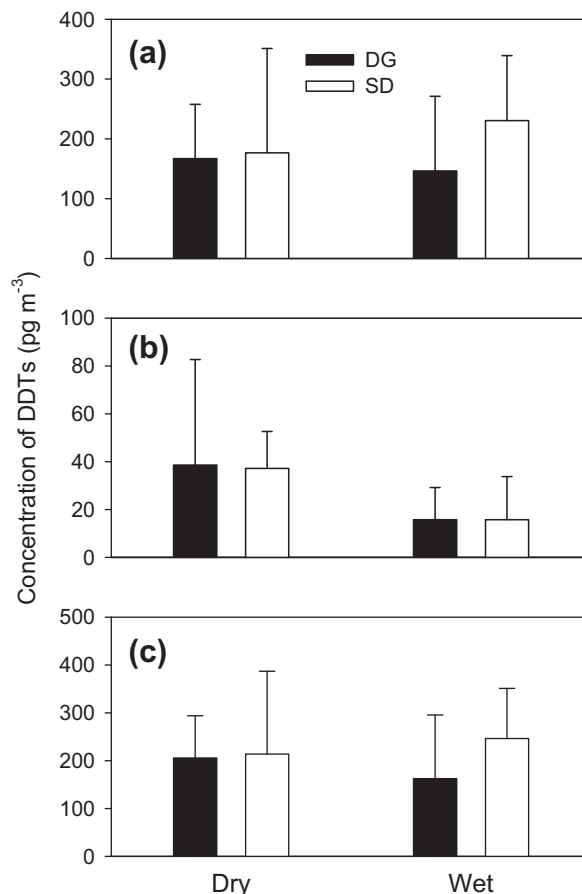

Fig. 2. Average concentrations of DDTs (sum of o,p'- and p,p'-DDT, DDE, and DDD) in (a) vapor phase; (b) particulate phase; and (c) air samples (gaseous + particulate phases) from Donguan (DG) and Shunde (SD) obtained in dry and wet weather seasons. The error bars are standard deviations of the concentrations.

120 pg m^{-3}) were lower than those from Shunde (240 ± 120 pg m^{-3}). Generally, the average concentration of DDTs in the air from Dongguan and Shunde (200 ± 120 pg m^{-3}) was much lower than those from both suburban and urban areas (mean: 1400 and 2000 pg m^{-3}, respectively) of Guangzhou (Fig. 1) reported by Li et al. (2007) and from the major cities (1470 ± 1010 pg m^{-3}) of India including New Deli, Kolkata, Mumbai, Chennai, Bangalore, Goa, and Agra (Chakraborty et al., 2010), comparable to those from Guangzhou (300 pg m^{-3}) by Yang et al. (2008), Tianjin (mean: 330 pg m^{-3}) (Zheng et al., 2010), and New Brunswick (mean: 247 pg m^{-3}) (Gioia et al., 2005), but higher than those in Camden, Delaware Bay, Jersey City, Pinelands and Sandy Hook in the Mid-Atlantic region of the United States (140, 41, 33, 33 and 22 pg m^{-3}, respectively) (Gioia et al., 2005).

In terms of dry weather season (from October to March) and wet weather season (from April to September) (Fig. 2), the average concentrations of gaseous DDTs from both Dongguan and Shunde were less variable (coefficients of variation (%CVs): 9.2% and 18.7%, respectively) than those of particulate DDTs (%CVs: 59.6% and 57.4%, respectively). However, no plausible explanations of these findings can be provided presently as the governing factors may have been comprehensive and complicated.

The dissolved VWM concentrations of DDTs in the rural areas of Dongguan and Shunde were 700 ± 150 and 670 ± 170 pg L^{-1}, respectively, both lower than that in the urban area in Guangzhou (1140 ± 500 pg L^{-1}) (Fig. 3). In addition, the particulate VWM con-centrations of DDTs in Dongguan and Shunde were 238 ± 66 and
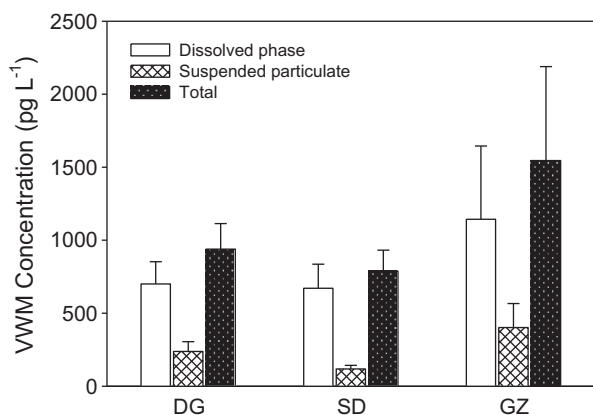
Fig. 3. Volume weighted mean (VWM) concentrations of DDTs in the dissolve and particle phases, and combined samples of precipitation from Donguan (DG), Shunde (SD) and Guangzhou (GZ).

118 ± 25 pg L^{-1}, respectively, also lower than that in Guangzhou (400 ± 160 pg L^{-1}) (Fig. 3). The total VWM concentrations of DDTs (dissolved plus particulate phases) in these three areas were higher than those in precipitation from Camden (190 ± 75 pg L^{-1}; $n = 12$), Jersey City (367 ± 184 pg L^{-1}; $n = 13$), and Pinelands (58 ± 19 pg L^{-1}; $n = 30$) in the mid-Atlantic region of the United States (Gioia et al., 2005), in precipitation (490 pg L^{-1}; sum of p,p'-DDT, p,p'-DDD and p,p'-DDE) from Shanghai (Xiong et al., 2007), and in Lake Malawi of southern Africa (76 ± 118 pg L^{-1}) (Karlsson et al., 2000).

The finding from the present study that levels of DDTs in both air and precipitation were higher in urban areas than in rural areas is consistent with previous results for atmosphere or other environmental compartments (Li et al., 2007; Wang et al., 2007; Ma et al., 2008; Yang et al., 2008; Liu et al., 2009). Although further evidence is needed to address this issue, recycling of legacy DDTs due to rapid urbanization in the PRD may be the main reason for the above observations. In essence, a large portion of DDTs historically used may have been applied in the surrounding rural areas of urban centers as the utility of DDTs was cost-prohibitive for most remote rural areas. The rural areas adjacent to large urban centers have gradually been urbanized as a result of the accelerating economic development since the early 1980s. Consequently, DDTs

sequestered historically could be recycled into the environment with conversion of agricultural lands into industrial and residential use, which has also expanded the territory of an urban center.

3.2. Phase partitioning in air and precipitation

It is well established that partitioning of a chemical between the atmosphere and organic phase in other environmental compartments such as soil, vegetation, and aerial suspended particles, described by the octanol–air partition coefficient (K_{oa}, mL g^{-1}) or between water and organic matter in a solid phase, depicted as the octanol–water partition coefficient (K_{ow}, mL g^{-1}), is a critical factor governing the fate, transport, and transformation of chemicals in the environment (Bidleman, 1988). Because GFF with a nominal pore size of 0.6 μm was used to collect air particles, only operationally defined partition coefficient between air and particulate organic matter (K'_{oa}, mL g^{-1}) can be calculated, which is expressed by

$$K'_{oa} = C_P/(TSP \cdot f_{om} \cdot C_G) = C'_P/(TSP \cdot 1.724 f_{oc} \cdot C_G) \quad (3)$$

where C_P and C_G (pg m^{-3}) are the particulate and gaseous concentrations of DDTs, respectively, TSP is the total suspended particulate concentration (g cm^{-3}), f_{om} and f_{oc} are the fractions of organic matter and organic carbon ($f_{om} = 1.724 \times f_{oc}$, (Sabljić, 1989; Schwarzenbach et al., 1993), respectively, in particles. The values of TSP and f_{oc} for each precipitation sample are listed in Tables S1 (Supplementary material).

Values of log K'_{oa} for p,p'-DDT, p,p'-DDE, p,p'-DDD, o,p'-DDT, o,p'-DDE, and o,p'-DDD were 9.64 ± 0.58, 10.07 ± 0.56, 9.90 ± 0.76, 10.06 ± 0.66, 10.02 ± 0.72, and 10.13 ± 0.57, respectively (Table 1). In comparison, Shoeib and Harner (2002) reported the log K_{oa} values of p,p'-DDT, p,p'-DDE, p,p'-DDD, and o,p'-DDT as 9.82, 9.68, 10.10, and 9.45. On the other hand, log K_{oa} values of p,p'-DDT, p,p'-DDE, p,p'-DDD, o,p'-DDT, o,p'-DDE, and o,p'-DDD estimated from the software of the Estimation Programs Interface (EPI) Suite of the United States Environmental Protection Agency (USEPA) are 10.38, 9.28, 9.59, 9.28, 9.35, and 9.35. Independent t-test showed no significant difference ($p > 0.05$) between log K'_{oa} and measured log K_{oa} or between log K'_{oa} and estimated log K_{oa}.

Similar to the phase partitioning in air, phase distribution in precipitation is somewhat influenced by colloids that enter into the dissolved phase through the GFFs with a nominal pore size of 0.7 μm used to collect particles in precipitation. As a result, only

Table 1
Operationally defined partition coefficients between the gaseous phase and particulate organic matter in air (K'_{oa}) and partition coefficients between the dissolved phase and particulate organic matter in precipitation (K'_{om}) of DDT components, particle associated fractions (φ) and particle-sorbed levels of DDT components (C_P, pg m^{-3}) in air, volume weighted mean concentrations of DDT components in precipitation (VWM, pg L^{-1}), and total washout ratios (W_T) for DDT components in Dongguan (DG), Shunde (SD) and Guangzhou (GZ).

		p,p'-DDT	p,p'-DDE	p,p'-DDD	o,p'-DDT	o,p'-DDE	o,p'-DDD
Log K'_{oa}		10.06	10.07	10.13	9.90	10.02	9.64
Log K'_{om}		6.37	6.36	6.14	6.01	6.49	6.58
φ	DG	0.27	0.14	0.50	0.23	0.30	0.11
	SD	0.12	0.16	0.12	0.48	0.18	0.32
	GZ[a]	0.13	0.12	~0	0.41	0.15	0.26
C_P	DG	2.0	7.1	1.9	2.5	3.5	2.9
	SD	3.3	3.5	4.4	1.7	6.0	3.6
	GZ[a]	2.8	3.6	nd	7.4	16	16
VWM	DG	46	280	66	150	160	240
	SD	65	230	33	170	73	220
	GZ	92	480	98	250	180	440
W_T	DG	10,000	10,000	19,000	16,000	9900	22,000
	SD	20,000	15000	54,000	5000	6700	9400
	GZ	8000	20,000	19,000	4600	8700	9700

[a] Particle associated fractions and particle-sorbed levels of DDT components in air were obtained from (Yang et al., 2008), nd = not detected. When calculating the W_T of p,p'-DDD in Guangzhou, particle-sorbed level of p,p'-DDD was replaced by half of the reported limit of detection at 1.6 pg m^{-3}.

the operationally defined partition coefficient between water and particulate organic matter (K'_{om}, mL g^{-1}) was calculated:

$$K'_{om} = C'_P/(TSP' \cdot f'_{om} \cdot C_D) = C'_P/(TSP' \cdot 1.724 f'_{oc} \cdot C_D) \qquad (4)$$

where C'_P and C_D (pg m^{-3}) are the particulate and dissolved phase concentrations of DDTs in precipitation, respectively, TSP' (g cm^{-3}) is the total suspended particulate concentration in precipitation, f'_{om} and f'_{oc} are the fractions of organic matter and organic carbon in particles of precipitation, respectively. The values of TSP' and f'_{oc} for each precipitation sample are listed in Tables S2 (Supplementary material).

The log K'_{om} values for p,p′-DDT, p,p′-DDE, p,p′-DDD, o,p′-DDT, o,p′-DDE, and o,p′-DDD were 6.58 ± 0.66, 6.36 ± 0.53, 6.01 ± 0.62, 6.41 ± 0.42, 5.98 ± 0.76, and 5.95 ± 0.66, respectively (Table 1). Correspondingly, the experimental values of log K_{ow} for these chemicals were 6.91, 6.96, 6.02, 6.79, 6.00, and 5.87, respectively (the first two values from Sabljic et al. (1995) and others from Howard and Meylan (1997)). The differences between log K'_{om} and log K_{ow} may reflect the impact of colloids on the quantification of phase partitioning, as well as the difference between octanol and natural organic matter as sorbent media for partitioning of organic chemicals. Independent t-test showed no significant difference ($p > 0.05$) between log K'_{om} and measured log K_{ow}. The applications of log K'_{oa} and log K'_{om} will be further discussed in the following section.

3.3. Washout ratios

Precipitation can washout organic chemicals in the gaseous or particulate phase from the atmosphere. The washout ratio W_T, defined as the ratio of a chemical's concentration in precipitation to its concentration in air, represents the efficiency of precipitation in eliminating chemicals from air, and can be calculated by

$$W_T = (1 - \varphi) \cdot W_G + \varphi \cdot W_P \qquad (5)$$

where W_G and W_P are the vapor and particle washout ratios, respectively, and φ is the particle-sorbed fraction in air (Wania and Westgate, 2008). Concentrations of DDTs in the gaseous phase of air and the dissolved phase of rain droplets can hardly be synchronously measured. However, diffusion of DDTs is generally considered to be sufficiently rapid so that equilibrium distribution between the gaseous phase and rain droplets is assumed, which implies that W_G equals to the operationally defined equilibrium water–air partition coefficient (K'_{wa}) (Venier and Hites, 2008). K'_{wa} can be calculated by the ratio of K'_{oa} to K'_{om} (Table 1). On the other hand, chemicals affiliated with particles in air are supposed to be adequately scavenged (Ligocki et al., 1985; Rounds et al., 1993); therefore, W_P equals to the ratio of the chemical concentration in precipitation to that in aerial particles (Table 1). With all these considerations, W_T is calculated by

$$W_T = (1 - \varphi) \cdot K'_{oa}/K'_{om} + \varphi \cdot VWM/C_P \qquad (6)$$

The annual mean W_T of individual DDT components were estimated at 9900–22,000, 5000–54,000, and 4600–20,000 in Dongguan, Shunde, and Guangzhou, respectively (Table 1). The W_T of p,p′-DDE acquired in the present study was more than double that measured in College Station of Texas, USA, but W_T of p,p′-DDT was less than half of the value in the previous study (Atlas and Giam, 1988). The difference between the two results may partly be attributed to the different methods applied in calculating W_G. Because gaseous phase and precipitation samples are difficult to obtain simultaneously, W_G is normally calculated based on gaseous phase concentrations obtained during non-raining periods (Atlas and Giam, 1988; Zhang et al., 2009), resulting in underestimated values of W_G. Furthermore, W_G values obtained by the methods of Atlas and Giam (1988) and Zhang et al. (2009) could be quite variable depending on the duration of sampling time in rain, i.e., longer sampling time leads to lower W_G (Koester and Hites, 1992; van Pul et al., 1999). On the other hand, W_G was calculated in the present study based on the operationally defined equilibrium partitioning between air and rain droplets, eliminating any potential negative impact from rainfall duration.

3.4. Atmospheric depositional intensity

Aerial deposition includes dry deposition of particles, rain/snow scavenging of vapors and particles, and gaseous exchange between the air-surface interface (Cousins et al., 1999). In the present study, air-surface gaseous exchange of DDTs could not be estimated due to insufficient data. The intensities of dry particle and wet deposition (I_D and I_W, respectively, g m^{-2} y^{-1}) are calculated by (Venier and Hites, 2008)

$$I_D = C_P \cdot v_d \qquad (7)$$

$$I_W = VWM \cdot p \qquad (8)$$

where v_d and p are the dry depositional velocity of particles in air (m y^{-1}) and annual precipitation rate (m y^{-1}), respectively. Particle dry depositional velocities depend largely on the physical properties of particles, such as particle size and density, and meteorological conditions, such as humidity and wind speed, and therefore often vary in a wide range. For example, Venier and Hites (2008) reported v_d in the range of 0.2–49 cm s^{-1}. An empirical value of 0.5 cm s^{-1} for v_d was selected to estimate the dry depositional fluxes of polybrominated diphenyl ethers in the study areas (Zhang et al., 2009), while Li et al. (2010) reported an average v_d value of 0.28 cm s^{-1} from dry particle deposition of BDE-47, BDE-99, and BDE-209 in Guangzhou. In the present study, 0.28 cm s^{-1} was chosen to calculate dry particle depositional intensities. Furthermore,

Table 2
Annual dry particle and wet depositional intensities of DDT components in Dongguan (DG), Shunde (SD), Guangzhou (GZ) (Fig. 1), and total depositional intensity of DDTs in the entire Pearl River Delta (PRD) compared with the Great Lakes (Buehler et al., 2003) and Hong Kong (Wong et al., 2004).

	p,p′-DDT	p,p′-DDE	p,p′-DDD	o,p′-DDT	o,p′-DDE	o,p′-DDD	DDTs
Dry particle depositional intensity (g m^{-2} y^{-1})							
DG	1.8×10^{-7}	6.3×10^{-7}	1.7×10^{-7}	2.2×10^{-7}	3.1×10^{-7}	2.6×10^{-7}	1.8×10^{-6}
SD	2.9×10^{-7}	3.1×10^{-7}	3.9×10^{-7}	1.5×10^{-7}	5.3×10^{-7}	3.2×10^{-7}	2.0×10^{-6}
GZ	2.5×10^{-7}	3.2×10^{-7}	7.1×10^{-8}	6.5×10^{-7}	1.4×10^{-6}	1.4×10^{-6}	4.1×10^{-6}
Wet depositional intensity (g m^{-2} y^{-1})							
DG	7.8×10^{-8}	4.8×10^{-7}	1.1×10^{-7}	2.6×10^{-7}	2.7×10^{-7}	4.1×10^{-7}	1.6×10^{-6}
SD	1.1×10^{-7}	3.9×10^{-7}	5.6×10^{-8}	2.9×10^{-7}	1.2×10^{-7}	3.7×10^{-7}	1.3×10^{-6}
GZ	1.6×10^{-7}	8.2×10^{-7}	1.7×10^{-7}	4.3×10^{-7}	3.1×10^{-7}	7.5×10^{-7}	2.6×10^{-6}
Total depositional intensity of DDTs (g m^{-2} y^{-1})							
PRD	Superior	Michigan	Huron	Erie	Ontario	Hong Kong	
3.7×10^{-6}	1.1×10^{-7}	2.0×10^{-7}	2.5×10^{-7}	1.1×10^{-7}	3.8×10^{-7}	nd[a]–7.8×10^{-8}	

[a] nd = not detected.

the average precipitation rate within the study areas in 2007 was $1.7 \, \text{m} \, \text{y}^{-1}$ (Zhang et al., 2009).

The dry particle depositional intensities of DDTs in the rural areas of Dongguan and Shunde and the urban area of Guangzhou were 1.8×10^{-6}, 2.0×10^{-6}, and $4.1 \times 10^{-6} \, \text{g} \, \text{m}^{-2} \, \text{y}^{-1}$, respectively (Table 2). In addition, the wet depositional intensities of DDTs in the rural areas of Dongguan and Shunde and the urban area of Guangzhou were 1.6×10^{-6}, 1.3×10^{-6}, and $2.6 \times 10^{-6} \, \text{g} \, \text{m}^{-2} \, \text{y}^{-1}$, respectively (Table 2). These results indicate that dry particle deposition is a slightly more predominant route for atmospheric deposition of DDTs in the PRD compared to wet deposition. If the present rural and urban data can be expanded to the entire PRD, depositional intensity for the entire PRD would be estimated. It has been noted that urban area covers approximately 10% of the total land in the PRD (Statistical Bureau of Guangdong Province, 2010), and the average intensities of dry particle and wet deposition would be 2.1×10^{-6} and $1.6 \times 10^{-6} \, \text{g} \, \text{m}^{-2} \, \text{y}^{-1}$, respectively. By contrast, the total depositional intensity in the PRD was much higher than that in Hong Kong (Wong et al., 2004) and those of Great Lakes areas (Buehler et al., 2003) (Table 2).

Acknowledgements

Appendix A. Supplementary material

Supplementary data associated with this article can be found, in the online version, at doi:10.1016/j.chemosphere.2011.03.045.

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