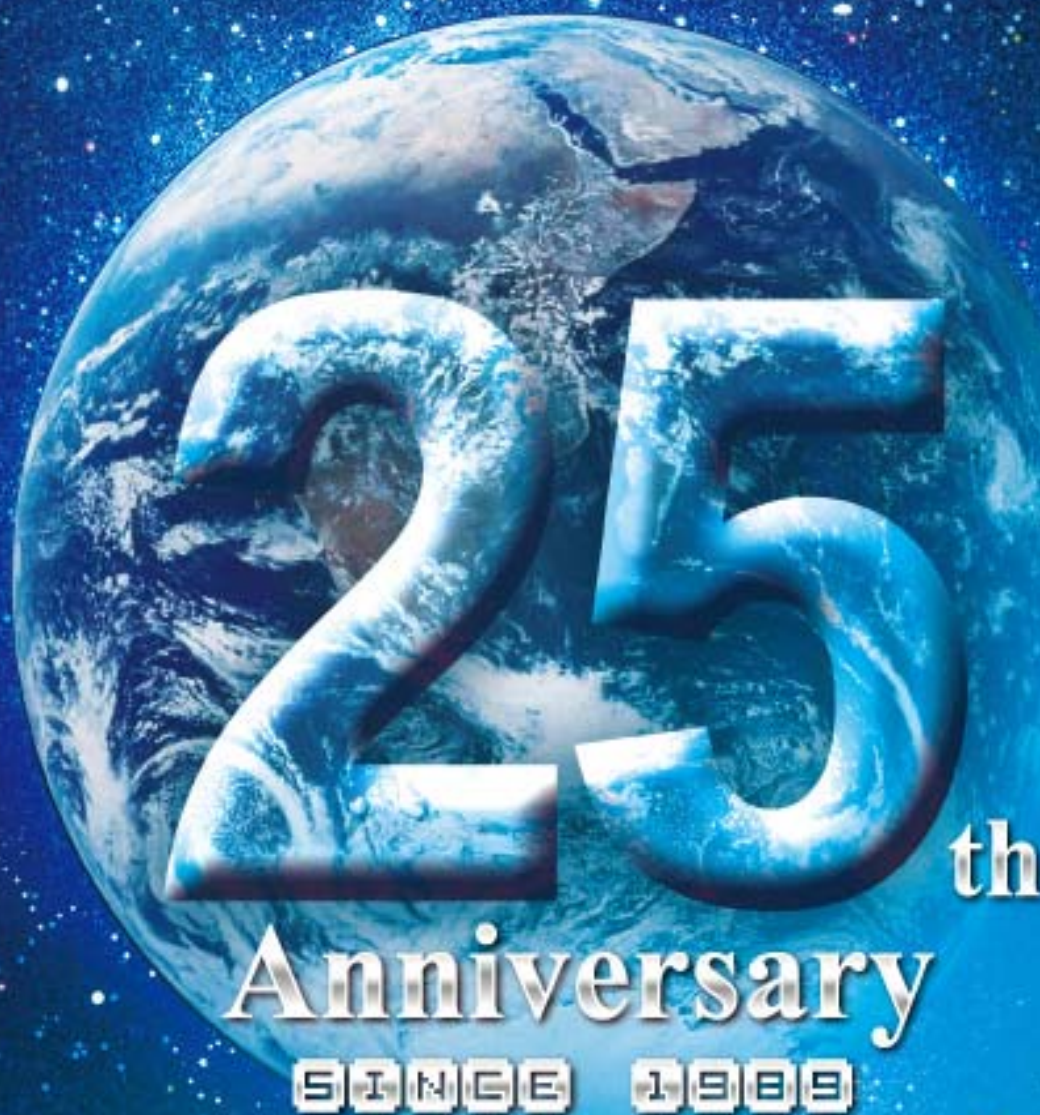


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## Simultaneous monitoring of PCB profiles in the urban air of Dalian, China with active and passive samplings

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### Abstract

The concentration of polychlorinated biphenyls (PCBs) in the urban air of Dalian, China was monitored from November 2009 to October 2010 with active high-volume sampler and semipermeable membrane device (SPMD) passive sampler. The concentration of PCBs (particle + gas) ( $\Sigma$ PCBs) ranged from 18.6 to 91.0  $\text{pg}/\text{m}^3$ , with an average of 50.9  $\text{pg}/\text{m}^3$ , and the most abundant dioxin-like PCB (DL-PCBs) was PCB118. The WHO-TEQ values of DL-PCBs were 3.6–22.1  $\text{fg}/\text{m}^3$ , with an average of 8.5  $\text{fg}/\text{m}^3$ , and PCB126 was the maximum contributor to  $\Sigma$ TEQ. There was a much larger amount of PCBs in the gas phase than in the particulate phase. The dominant PCB components were lower and middle molecular weight PCBs. With increasing chlorination level, the concentration of the PCB congeners in the air decreased. The gas-particulate partitioning of PCBs was different for the four seasons. The gas-particulate partitioning coefficients ( $\log K_p$ ) vs. subcooled liquid vapor pressures ( $\log P_L^0$ ) of PCBs had reasonable correlations for different sampling sites and seasons. The absorption mechanism contributed more to the gas-particulate partitioning process than adsorption. Correlation analysis of meteorological parameters with the concentration of PCBs was conducted using SPSS packages. The ambient temperature and atmospheric pressure were important factors influencing the concentration of PCBs in the air. The distribution pattern of the congeners of PCBs and the dominant contributors to DL-PCBs and TEQ in active samples and SPMDs passive samples were similar. SPMD mainly sequestered gas phase PCBs.

**Key words:** air; polychlorinated biphenyls; gas-particulate partitioning; semipermeable membrane device; Dalian; China

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### Introduction

Polychlorinated biphenyls (PCBs) are ubiquitous in the atmosphere and an important class of persistent organic pollutants, with the properties of persistence, toxicity, bioaccumulation and long-distance migration. They are seriously threatening both ecosystems and human health. Although banned for more than 30 years, PCBs are still detected globally in the environment (Kim et al., 2011). Long-range transport spreads PCBs worldwide and through the food chain to enter, either directly or indirectly, into the human body (Xing et al., 2005). Prior to 1974, PCBs generally entered the environment during their manufacture and use. In China, approximately 10,000 tons of PCBs were produced from 1965 to 1974 (Zhang et

al., 2008). Trichlorobiphenyl was used primarily in power capacitors applied in electricity production, distribution and transmission, while pentachlorobiphenyl was used mainly as a paint additive (China SEPA, 2003). There have been numerous reports on the PCB research of China in air, soil, water, sediments and aquatic organisms (Chen et al., 2008; Choil et al., 2008; Li et al., 2004, 2007; Meng et al., 2000; Sun et al., 2009; Xing et al., 2005; Zhu et al., 2009).

It is well known that the concentrations of PCBs in the air can be monitored by active and passive sampling techniques. These two methods each have advantages and disadvantages. With active sampling technique the actual concentration of PCBs in the air can be obtained directly, but the major drawbacks are the high costs of equipment, the need for a power supply and for maintenance by qualified persons. Passive sampling technology has be-

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come of great importance in the field of environmental monitoring, due to its unique advantages for long-term on-site monitoring of pollutants and the accumulation of chemicals via diffusion or sorption, with no electricity or any other power sources required to operate (Kot-wasik et al., 2007). Another important advantage of the integrative uptake achieved over long deployment periods is the detection of compounds otherwise unnoticed by active air sampling over a short period (Levy et al., 2008). But with passive sampling only the amount of PCBs sequestered in the device can be determined, while a main community concern is the actual concentration of PCBs in the air. The semi-permeable membrane device (SPMD) is one of the most popular passive samplers for sampling PCBs in the air (Esteve-Turrillas et al., 2008; Huckins et al., 2006; Zhu et al., 2007). The actual concentration of PCBs in the air can be obtained by the concentrations sequestered by SPMD, the sampling rate of SPMD and its exposure time (Lohmann et al., 2001; Zhu et al., 2011). Dalian (120°58'–123°31'E, 38°43'–40°10'N) is a seaside city located on the southern tip of the Eastern Liaoning Peninsula of northeast China. The petrochemical industry, modern equipment manufacturing, ship building, and electronic information products industry are the four pillar industries of Dalian. Although Dalian is one of the most livable cities in China, there are also various kinds of dispersed sources releasing PCBs that affect the air quality, such as motor vehicle exhausts, industrial emissions, heating, waste incineration, etc.

To our knowledge, there has been no relevant report on the simultaneous monitoring of the urban air PCBs of Dalian, China with active high-volume sampling method and SPMD passive sampling method. The present study is therefore devoted to this goal. The other aims of this study were to investigate the gas-particle distribution of PCBs in the air and the relationship of their concentrations to meteorological parameters, as well as the correlation between the concentrations of PCBs sequestered by SPMDs and those measured by active sampling.

## 1 Materials and methods

### 1.1 Sampling

The sampling process was conducted according to US EPA compendium method TO 9A. From November 2009 to October 2010 (except February 2010, the Chinese Spring Festival holiday), active high-volume air samplings (ECHO HiVol, TCR TECORA, Italy) were carried out at two sites in Dalian, China, which were selected based on their different surrounding environments, populations and traffic densities. The sampling sites were the rooftops of the No.1 building at Dalian Jiaotong University (DJU: 121°54.399'E, 38°34.286'N, elevation of 32 m) and the building of Dalian Meteorological Observatory (DMO: 121°38.730'E, 38°54.174'N, elevation of 113 m). All

sampling sites were located in open areas, without trees, buildings or any other sheltering objects near them.

DMO is located on a hillside. The surroundings of this site include residential buildings, university campus, a light traffic road and a vast expanse of mountain with many trees and plants on it. Dalian Port (DLP) is a world-known deep and ice-free harbor, having shipping relations with over 160 countries and regions. Dalian Newport (DNP) is a modern deepwater oil port. DMO is about 3 km from DLP and about 24 km from DNP.

DJU is located in the campus of Dalian Jiaotong University. It is close to residential buildings, research institutes, the university's restaurants, barbecue Stall Street, two heavy traffic roads and a coal-fired power plant at the southwest of DJU. DJU is about 8.2 km from DLP and about 29.9 km from DNP.

All samples were collected with a suction flow of 0.3 m<sup>3</sup>/min. Approximately 600 m<sup>3</sup> air was collected per sample. The particulate fraction was collected by a glass microfiber filter (GFF) (Whatman grade GF/A 110 mm diameter) and the gas phase was collected by a polyurethane foam (PUF) plug (length 76 mm, diameter 63 mm, density 0.0213 g/m<sup>3</sup>). The preconditioning of GFF and PUF was carried out as described by Wang et al. (2011). The air volume recorded by the sampler was used to calculate the concentrations of PCBs. The background contamination was monitored by using operational blanks (unexposed filter and PUF) which were processed simultaneously with field samples.

In this work, the SPMD was designed as a low-density polyethylene flat tubing (length 23 cm, width 2.5 cm, thickness  $\approx$  67.5  $\mu$ m) filled with 0.7 mL 99% triolein (Sigma-Aldrich, Taufkirchen, Germany). The area of each SPMD was about 115 cm<sup>2</sup>. The SPMDs were deployed in a Stevenson Screen of untreated wood (46 cm  $\times$  46 cm  $\times$  61.2 cm). Then, they were placed 1.5 m above the ground at the two sampling sites.

### 1.2 Meteorological conditions

Dalian Meteorological Observatory provided meteorological data synchronously with the sampling period. The parameters included annual average temperature, average pressure, average relative humidity, average wind speed and the concentrations of the total suspended particles (TSP). The meteorological data and TSP values are listed in Table S1.

### 1.3 Sample extraction and analysis

Actively generated samples were prepared according to US EPA method 1668A. All <sup>13</sup>C<sub>12</sub>-labeled PCB standards were obtained from Cambridge Isotope Laboratories Inc. USA (purities >98%). In general, the solvents and reagents used for the study were all pesticide grade or higher.

The GFFs and PUFs samples were separately transferred into Soxhlet glass thimbles, spiked with 12 congeners

of  $^{13}\text{C}_{12}$ -labelled PCB internal standards (i.e., PCB77, 81, 126, 169, 105, 114, 118, 123, 156, 157, 167, 189), and extracted for 18 hr with 250 mL *n*-hexane and dichloromethane (1:1, V/V). The sample cleanup was performed with a multi-layer silica gel column and alumina column. The final clean extract was stepwisely transferred into a micro-vial and evaporated to dryness with a gentle flow of nitrogen. Finally, 10  $\mu\text{L}$  of nonane containing 5 congeners of  $^{13}\text{C}_{12}$ -labelled recovery standard (i.e., PCB9, 52, 101, 138, 194) was added into the micro-vial for high resolution gas chromatography/high resolution mass spectrometry analysis.

Passive sampling samples were prepared as described by Zhu et al. (2007).

Active sampling samples were analyzed in the Dalian Institute of Chemical Physics, Chinese Academy of Sciences, Dalian, China. Passive sampling samples were analyzed in Helmholtz Zentrum München – German Research Center for Environmental Health, Institute of Ecological Chemistry, Germany. Instrumental analysis parameters are summarized in **Table 1**. Toxic equivalents (TEQ) values were calculated using 1998 World Health Organization toxicity equivalency factors (WHO-TEF).

#### 1.4 Quality assurance and quality control

Pre-cleaned GFF and PUF plugs were used for the particulate and gas phase sampling. All of the active sampling samples were spiked with labeled compounds to monitor the recovery performance of the method. Field blanks and laboratory blanks were incorporated in the analytical procedure for the purpose of quality control. There were no apparent differences between field blanks and laboratory blanks. Compared with the air samples, the target contaminants detected in the blank samples could be ignored. The detection limits were defined as three times the signal/noise ratio. The concentrations of the main interferences in blank samples were less than 5% of those in the air samples. The recovery efficiency for twelve

$^{13}\text{C}_{12}$ -labelled PCBs ranged from 54% to 138%, which was in compliance with US EPA method 1668A (25%–150%). The data were not recovery corrected.

The quality assurance and quality control of passive sampling was performed as described by Zhu et al. (2007). The data were corrected by solvent blanks.

## 2 Results and discussion

### 2.1 PCBs levels in the urban air of Dalian

A total of 86–109 and 64–107 congeners of PCBs were detected in the gas phase (PUF) and particulate phase (GFF) in the urban air of Dalian, respectively. The average total concentration of PCBs ( $\Sigma\text{PCBs}$ ) in PUF and GFF samples ranged from 16.8 to 87.8  $\text{pg}/\text{m}^3$  with an average of 41.8  $\text{pg}/\text{m}^3$  and 1.2 to 24.4  $\text{pg}/\text{m}^3$  with an average of 9.1  $\text{pg}/\text{m}^3$ , respectively. The WHO-TEQ of the total PCBs in PUF and GFF samples ranged from 1.5 to 6.3  $\text{fg-TEQ}/\text{m}^3$  with an average of 3.6  $\text{fg-TEQ}/\text{m}^3$  and 0.52 to 17.9  $\text{fg-TEQ}/\text{m}^3$  with an average of 4.9  $\text{fg-TEQ}/\text{m}^3$ , respectively. The total concentration (GFF + PUF) and WHO-TEQ of PCBs in the air of Dalian are listed in **Table 2**.

The average concentration of PCBs in the gas phase accounted for 82.1% of the total concentration, which was much higher than that of PCBs in the particulate phase. This indicated that PCBs were more likely to exist in a gaseous state in the atmosphere. Similar results were obtained by previous studies (Kim et al., 2011; Mandalakis et al., 2002; Simcik et al., 1998; Tasdemir et al., 2004, 2005; Totten et al., 2002).

The components of gas and particulate phase PCBs in the air of Dalian were similar.  $\Sigma\text{PCBs}$  were both dominated by the lower molecular weight species (tri-, tetra- and penta-PCBs), which accounted for >80% of the total concentration, while the ratio of the concentration of higher molecular weight species (hexa- to deca-PCBs) to the total concentration in the particulate phase was higher than that

**Table 1** Gas chromatography/mass spectrometry (GC/MS) parameter for the isomer specific detection of PCBs

	Active sample	Passive sample
Gas chromatography	Type: Hewlett-Packard 6890 Column: Rtx-5MS (Restec Company), 60 m, 0.25 mm ID, 0.25 $\mu\text{m}$ df film thickness Temperature program: 90°C, 2 min, 20°C/min, 180°C, 15 min, 3.0°C/min, 300°C, 30°C/min, 320°C, 10 min; Carrier gas: helium; Injection port temperature: 280°C; Temperature transfer line: 250°C; Injection volume: 1 $\mu\text{L}$ splitless;	Type: Agilent 5890 Series II; Column: ZB-MultiResidue2, 30 m, 0.25 mm ID, 0.2 $\mu\text{m}$ film thickness (Phenomenex); Temperature program: 90°C, 1.5 min, 20°C/min, 170°C, 7.5 min, 3.5°C/min, 265°C, 20°C/min, 310°C, 10 min; Carrier gas: helium, head pressure: 16 psi; Injector: Cold injection system CIS 3 (Gerstel); Temperature program injector: 120°C, 12°C/sec, 300°C, 5 min; Temperature transfer line: 310°C; Injection volume: 0.8 $\mu\text{L}$ splitless
Mass spectrometry	Type: Autospec Ultima HRMS (Micromass, UK); Ionization mode: EI, 37 eV; Ion source temperature: 250°C; Resolution: $\geq 10,000$ ; Detection: SIM mode.	Type: MAT 95 (Thermo); Ionization mode: EI, 47 eV; Ion source temperature: 260°C Resolution: > 8000; Detection: SIM mode

**Table 2** Concentrations (pg/m<sup>3</sup>), WHO-TEQs (fg-TEQ/m<sup>3</sup>) of PCBs,  $\Sigma$ DL-PCBs,  $\Sigma$ PCBs and  $n$  in the urban air of Dalian

DJU sampling date		2009.11	2009.12	2010.01	2010.03	2010.04	2010.05	2010.06	2010.07	2010.08	2010.09	2010.10
Indicator-PCBs	PCB28	6.103	1.931	6.313	2.620	2.719	2.627	4.454	6.567	5.779	5.980	1.236
	PCB52	1.044	0.769	1.449	0.514	0.740	1.003	1.036	3.286	1.665	1.525	0.356
	PCB101	1.126	1.741	2.308	0.304	1.012	1.374	0.714	5.469	1.213	1.281	0.431
	PCB138	0.784	1.788	1.869	0.191	0.226	0.221	0.228	0.551	0.416	0.404	0.164
	PCB153	0.251	0.283	0.327	0.219	0.213	0.292	0.234	0.483	0.436	0.388	0.144
	PCB180	0.458	0.227	0.559	0.121	0.145	0.136	0.120	0.205	0.177	0.182	0.082
Non-ortho PCBs	PCB77	0.393	0.395	0.787	0.213	0.205	0.169	0.157	0.260	0.251	0.359	0.126
	PCB81	0.088	0.111	0.198	0.037	0.051	0.005	0.036	0.076	0.061	0.080	0.032
	PCB126	0.135	0.164	0.360	0.054	0.054	0.030	0.016	0.027	0.031	0.058	0.036
	PCB169	0.064	0.051	0.086	0.014	0.019	0.014	0.010	0.189	0.014	0.022	0.015
Mono-ortho PCBs	PCB105	0.561	0.610	0.764	0.380	0.358	0.430	0.327	0.601	1.089	0.633	0.312
	PCB114	0.057	0.070	0.128	0.036	0.031	0.040	1.288	0.059	0.055	0.123	0.034
	PCB118	1.178	1.420	1.687	0.875	0.789	1.036	0.902	1.697	1.741	1.666	0.277
	PCB123	0.070	0.062	0.111	0.038	0.166	0.043	0.000	0.066	0.000	0.000	0.000
	PCB156	0.201	0.057	0.181	0.114	0.149	0.146	2.489	0.188	0.177	0.193	0.112
	PCB157	0.050	0.077	0.017	0.027	0.039	0.039	1.909	0.051	0.031	0.052	0.029
	PCB167	0.185	0.142	0.343	0.072	0.080	0.075	0.051	0.157	0.100	0.117	0.049
	PCB189	0.101	0.089	0.199	0.032	0.029	0.018	0.014	0.018	0.014	0.033	0.024
$\Sigma$ DL-PCBs		3.083	3.248	4.860	1.894	2.996	2.088	7.207	3.212	3.112	3.282	1.505
$\Sigma$ PCBs		46.024	35.617	65.671	25.050	26.685	28.359	36.139	79.044	42.790	46.685	12.295
$n$ (PUF/GFF)		94/96	100/107	102/105	103/99	94/93	93/86	95/69	95/90	95/71	97/95	97/87
$\Sigma$ WHO-TEQ		14.587	17.320	37.425	5.752	5.854	3.438	5.470	3.251	3.682	6.458	3.942
DMO sampling date		2009.11	2009.12	2010.01	2010.03	2010.04	2010.05	2010.06	2010.07	2010.08	2010.09	2010.10
Indicator-PCBs	PCB28	4.582	5.041	3.969	13.841	7.716	5.773	6.492	11.237	6.388	3.091	3.094
	PCB52	0.889	1.460	1.537	1.088	2.280	2.286	3.437	2.713	1.989	0.781	0.733
	PCB101	0.905	1.438	1.909	0.587	15.338	3.609	2.691	4.620	2.212	0.784	0.618
	PCB138	1.046	1.686	0.915	0.279	0.524	0.306	0.561	0.855	0.464	0.179	0.270
	PCB153	0.333	0.263	0.263	0.293	0.553	0.380	0.645	0.822	0.400	0.189	0.264
	PCB180	0.271	0.218	0.193	0.122	0.451	0.144	0.142	0.348	0.344	0.111	0.147
Non-ortho PCBs	PCB77	0.317	0.399	0.196	0.432	0.377	0.222	0.306	0.475	0.340	0.203	0.226
	PCB81	0.057	0.082	0.045	0.067	0.108	0.071	0.118	0.182	0.083	0.038	0.073
	PCB126	0.075	0.112	0.064	0.057	0.097	0.036	0.040	0.084	0.030	0.023	0.111
	PCB169	0.016	0.031	0.015	0.012	0.026	0.017	0.011	0.034	0.006	0.009	0.041
Mono-ortho PCBs	PCB105	0.566	0.531	0.331	0.553	1.076	0.477	1.242	1.488	0.714	0.298	0.459
	PCB114	0.045	0.054	0.033	0.054	0.147	0.055	0.122	0.142	0.034	0.033	0.063
	PCB118	1.408	1.301	0.791	1.266	2.606	1.328	3.612	4.080	2.066	0.784	1.117
	PCB123	0.051	0.027	0.034	0.057	0.110	0.066	0.000	0.506	0.000	0.000	0.000
	PCB156	0.079	0.036	0.055	0.099	0.490	0.202	0.228	0.464	0.127	0.077	0.220
	PCB157	0.040	0.034	0.044	0.039	0.119	0.054	0.042	0.114	0.033	0.020	0.063
	PCB167	0.139	0.130	0.143	0.101	0.194	0.137	0.138	0.723	0.086	0.045	0.087
	PCB189	0.030	0.054	0.028	0.021	0.074	0.024	0.016	0.041	0.014	0.014	0.074
$\Sigma$ DL-PCBs		2.823	2.790	1.779	2.758	5.423	2.690	5.874	8.334	3.533	1.545	2.534
$\Sigma$ PCBs		41.295	54.566	51.077	89.837	86.980	68.798	74.702	103.000	56.717	23.656	24.931
$n$ (PUF/GFF)		109/107	105/104	104/97	102/98	86/91	93/90	97/84	94/87	97/64	97/92	96/94
$\Sigma$ WHO-TEQ		7.956	11.793	6.717	6.153	10.760	4.177	4.785	9.808	3.458	2.604	11.853

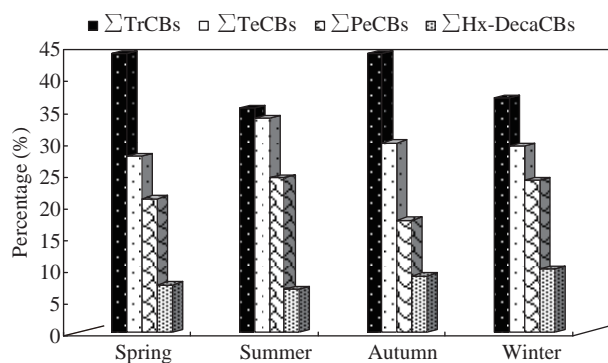
$n$  (PUF/GFF): the total number of detected congeners of PCB in gas phase and particulate phase, respectively.

in the gas phase. This was in agreement with the previous studies (Harner et al., 1998; Yeo et al., 2003). In general, the vapor pressures of the lower chlorinated PCBs are higher than those of the higher chlorinated PCBs. Lower chlorinated PCBs can readily evaporate from, e.g., soil into the atmosphere and the higher chlorinated PCBs exhibit higher affinity toward the particulate matter. Therefore, the contents of the lower chlorinated PCBs were larger in the gas phase, while those of the higher chlorinated PCBs were larger in the particulate phase.

**Table S2** is a comparison of concentrations and WHO-TEQ values of PCBs in the urban air of Dalian with other

cities in the world. Their concentration was quite low in comparison with those of other cities around the world. The TEQ level of the total dioxin-like PCBs (DL-PCBs) in the Dalian atmosphere was close to Japan (Kim et al., 2005), but higher than Turkey (Cindoruk et al., 2007).

Seasonal variations of the relative average concentrations of PCBs with different chlorinated atoms are shown in **Fig. 1**. Although the distribution patterns of the PCBs congeners among the four seasons (Winter: Dec 2009–Jan 2010; Autumn: Sep–Oct 2010, Nov 2009; Summer: June–Aug, 2010; Spring: March–May 2010) had some differences, similar variation regularity was observed. In



**Fig. 1** Seasonal variations of the relative concentrations of different chlorinated PCBs.

general, the concentration of the congeners decreased with the increasing number of chlorine atoms. However, the reason for the relatively lower proportion of tri-PCBs in summer is not very clear. The reason might be that the ambient temperature was higher in the summer and since the vapor pressure of tri-PCBs was also higher, they could more easily evaporate and transport over long distances, so the local concentration of tri-PCBs was lower. The dominant components were the lower chlorinated PCBs (tri-, tetra- and penta-PCBs), which accounted for 90.0%–93.2% of  $\Sigma$ PCBs in the urban air of Dalian. Li et al. (2007) also found that tri- and tetra-PCBs showed the greatest contribution in the atmosphere of the urban, rural and background areas of the Pearl River Delta in winter, and both contribution rates were very similar between different regions; Chen et al. (2008) indicated that the PCBs were dominated by tri-, tetra- and penta-PCBs in the Guangzhou atmosphere in summer, as well. However, these distribution patterns of the PCBs congeners were quite different from those of many other foreign cities. Duinker et al. (1989) found that penta- and hexa-PCBs were dominant in the atmosphere of the western Baltic. Brunciak et al. (2001) found that tetra- and penta-PCBs were the most abundant in the atmosphere of the Chesapeake Bay, USA. One of the reason might be that the production and use of PCBs were mainly based on trichloro biphenyl in China (Chen et al., 2007), while relatively higher chlorinated PCBs products were used in the foreign industries, such as Aroclor1254 and Aroclor1260 (Schulz et al., 1989; Taniyasu et al., 2003).

## 2.2 Distribution of DL-PCBs in the gas-particulate phases

Persistent organic pollutants in the global migration and transformation process are affected by many factors, such as vapor pressure, degradation and gas-particulate distribution dynamics, etc. A key process in determining the environmental fate of persistent organic pollutants in the atmosphere is their partitioning between particulate and gas phases (Oh et al., 2001). PCBs consist of 209 chemical compounds, among which, 12 PCBs (8 mono-ortho PCBs: PCB105, 114, 118, 123, 156, 157, 167 and 189; 4 non-

ortho PCBs: PCB77, 81, 126 and 169) have toxicity similar to that of dioxins which are assigned WHO-TEF. Hence, these compounds are referred to DL-PCBs.

It is well-known that the sub-cooled vapor pressure ( $P_L^0$ ) and the octanol-air partitioning coefficient ( $K_{OA}$ ) of semi-volatile organic compounds have good correlation with their gas-particle partitioning coefficient ( $K_p$ ) ( $\log K_p = m_r \log P_L^0 + b_r$ ;  $\log K_p = m \log K_{OA} + b$ ), and the partitioning of semi-volatile organic compounds between the gas and particulate phases has been described by Junge-Pankow and  $K_{OA}$ -based models (Finiaio et al., 1997; Harner and Bidleman, 1998; Junge, 1997; Lohmann et al., 2000b; Pankow, 1987, 1994).

The partitioning is believed to be governed by absorption and adsorption, and the former is more relevant to organic carbon content, while the latter is often associated with the surface area and black carbon content (Goss and Schwarzenbach, 1998). It is usually described by  $K_p$  (Hart et al., 1992; Yamasaki et al., 1982):

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

where,  $F$  ( $\text{pg}/\text{m}^3$ ) and  $A$  ( $\text{pg}/\text{m}^3$ ) are the concentrations of PCBs in the particulate and gas phases, respectively, and  $TSP$  ( $\mu\text{g}/\text{m}^3$ ) is the concentration of total suspended particulate matter. According to the above definition, the compounds with larger  $K_p$  values will show a tendency to adhere preferentially to atmospheric particles. The  $K_p$  values of indicator-PCBs and DL-PCBs in different seasons and the whole year were calculated according to Eq. (1) using the field data. Only the detected data of the analytes above the detection limits in both phases were included in the calculation. Generally, the seasonal variations of the  $\log K_p$  values of PCBs at the two sampling sites were winter > spring > or  $\approx$  autumn > summer.

**Table 3** shows the regressions of  $\log K_p$  versus  $\log P_L^0$  at the two sampling sites during the four seasons and the whole year, respectively. Temperature-dependent vapor pressure for indicator-PCB and DL-PCB congeners were calculated using  $m_L$  and  $b_L$  ( $\log P_L^0 = m_L/T + b_L$ ) data given by Falconer and Bidleman (1994) and adjusted to the average ambient temperature during the sampling periods.

For equilibrium partitioning of the chemicals between the gas and particulate phases, the slope  $m_r$  for either adsorption or absorption should be close to  $-1$  (Pankow, 1994). However, Goss and Schwarzenbach (1998) and Simcik et al. (1998) argued that a slope value of  $-1$  in the regression of  $\log K_p$  versus  $\log P_L^0$  was not necessary to describe equilibrium partitioning. Slopes significantly steeper than  $-1$  indicated adsorption and slopes shallower than  $-0.6$  pointed to absorption, while slopes in the range between  $-0.6$  and  $-1$  could occur for absorption as well as for adsorption (Goss and Schwarzenbach, 1998). The intercept  $b_r$  was dependent on the type and properties associated with the aerosol.

From **Table 3**, it can be seen that the slope values



**Table 3** Correlations of  $\log K_p$  versus  $\log P_L^0$ 

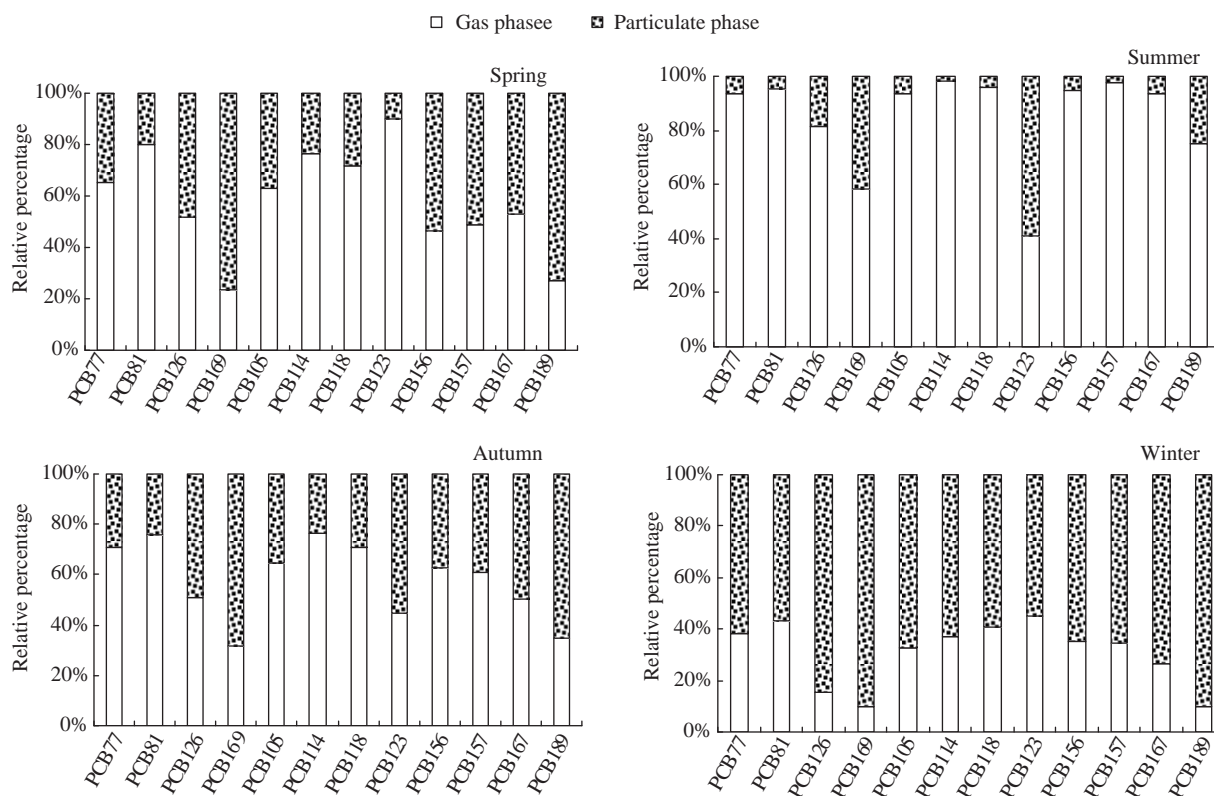
Sampling site	Parameter	Spring	Summer	Autumn	Winter	Whole year
DMO	$r^2$	0.59	0.63	0.79	0.49	0.61
	$T$ ( $^{\circ}\text{C}$ )	10.8	24.0	9.2	-6.4	10.7
	$m_r$	-0.44	-0.50	-0.32	-0.15	-0.34
	$b_r$	-3.85	-4.55	-3.47	-2.03	-3.58
DJU	$r^2$	0.90	0.30	0.71	0.70	0.40
	$T$ ( $^{\circ}\text{C}$ )	7.0	21.9	13.8	-4.4	10.9
	$m_r$	-0.51	-0.40	-0.43	-0.65	-0.42
	$b_r$	-4.28	-4.46	-3.87	-4.34	-3.85

were  $> -1$ , indicating that PCBs did not approach equilibrium between the particulate and gas phases. The slope values were shallower than  $-0.6$  (except at DJU in the winter season), indicating that the absorption mechanism contributed more to the partitioning process (Goss and Schwarzenbach, 1998). The  $m_r$  and  $b_r$  at both sampling sites for the whole year were similar, but they were different at the same sampling site in the four seasons. Other researchers (Kim et al., 2005, 2011; Lohmann et al., 2000b; Mandalakis et al., 2002; Tasdemir et al., 2004) also found that  $m_r$  and  $b_r$  showed large variations from location to location and event to event.

No matter whether in the gas phase or in the particulate phase, the monthly average total concentration variation of DL-PCB and indicator-PCB congeners at the two sampling sites were similar (Table 2) and when all data of gas phase or particulate phase samples at DJU and DMO were compared by the  $t$ -Test, the concentrations of DL-

PCBs and indicator-PCBs at the two sampling sites were not significantly different ( $P > 0.05$ ), so the average concentrations of DL-PCBs and indicator-PCBs at the two sampling sites were used in the following discussion.

The relative concentration distribution of DL-PCBs congeners in different seasons are shown in Fig. 2. During spring and autumn, the distributions of the average concentrations of DL-PCBs in gas-particulate phases were similar, the DL-PCB congeners were mainly in the gas phase, with an average contribution of 66.1% and 66.3% to the total concentrations in spring and autumn, respectively. During the summer, the average contribution of DL-PCB congeners in the gas phase was 94.1% of the total concentration, while in the winter, the DL-PCB congeners in the particulate phase accounted for 64.8% of the total concentrations. The heating period of Dalian was from 15, November to the end of March of the next year. Coal was the main fuel used for central heating in Dalian, so

**Fig. 2** Relative concentration distribution of DL-PCB congeners in the four seasons.

there were larger concentrations of particulates in the air in winter. The lower temperature enhanced the enrichment of PCBs in the particulate phase. Meanwhile, the higher temperature was conducive to the volatilization of PCBs into the atmosphere, resulting in a higher concentration of gas phase PCBs in the summer.

The major congeners of DL-PCB through the whole year were PCB118, 105 and 77, and their relative mean concentrations in the gas and particulate phases were 47.7%, 17.0%, 8.3% and 38.3%, 19.4%, 10.8%, respectively. This distribution pattern was similar to that of DL-PCBs in solid waste incineration flue gas (Chi et al., 2006), the PCB products in Japan, such as the rust remover KC-200, KC-300 and KC-400 (Takasuga et al., 2005), and was consistent with those reported in other studies on DL-PCBs (Castro-Jimenez et al., 2008; Choi et al., 2008; Helm and Bidleman, 2005; Kim et al., 2011). The main congeners of PCBs in the domestic PCB products were PCB105 and 77; for example, 1<sup>#</sup>PCB product contained 42% chlorine and trichloro biphenyl dominated, which was mainly used for power capacitor impregnant. 2<sup>#</sup>PCB product contained 53% chlorine and pentachloro biphenyl dominated, which was mainly used as a paint additive. In 1<sup>#</sup>PCB and 2<sup>#</sup>PCB products, the ratios of PCB105 and 77 were 19%, 81% and 96%, 2%, respectively, but PCB118 was not detected (Jiang et al., 1997). Thus, the results of this study were not consistent with the distribution patterns of PCBs in the power capacitors of China. This means that PCBs in the transformer oil made in China were not the only source of PCBs in the urban air of Dalian.

The average contribution rates of the TEQ of  $\Sigma$ DL-PCBs in the gas phase to the total TEQ were 51.8%, 83.7%, 50.8% and 15.5% during spring, summer, autumn and winter, respectively. PCB126 had the highest TEQ value among all the DL-PCBs in the gas and particulate phases, and it accounted for over 86.9% and 93.8% of the total TEQ in the above two phases, respectively. Yu et al. (2007) reported that the TEQ value of PCB126 accounted for 64% in the atmospheric particulates of the transformer and capacitor dismantling area of China. A maximum contribution of PCB126 to  $\Sigma$ TEQ was also found in Japan (Sakai et al., 2001), South Korea (Kim et al., 2007, 2011) and UK (Lohmann et al., 2000a).

### 2.3 Changes of PCBs concentration with spatial variation

A total of 93–103 and 69–107 congeners of PCBs were detected in the gas phase and particulate phase at DJU, respectively. The concentrations of total PCBs in gas and particulate phases at DJU were 10.9–77.2 pg/m<sup>3</sup> with an average of 32.5 pg/m<sup>3</sup> and 0.93–26.5 pg/m<sup>3</sup> with an average of 7.9 pg/m<sup>3</sup>, respectively. A total of 86–109 and 64–107 congeners of PCB were detected in the gas and particulate phases at DMO, respectively. The concentration of total PCBs in the gas and particulate phases at DMO

were 22.3–98.4 pg/m<sup>3</sup> with an average of 51.1 pg/m<sup>3</sup> and 0.99–42.4 pg/m<sup>3</sup> with an average of 10.4 pg/m<sup>3</sup>, respectively. Detailed data can be found in **Table 2**.

The monthly concentration of the total PCBs in the air at DMO was generally higher than that at DJU (**Table 2**). However, the TEQ values of  $\Sigma$ DL-PCBs at DJU were higher than those at DMO in the winter and the highest TEQ value of  $\Sigma$ DL-PCBs was found at DJU in January 2010. January was the coldest time in Dalian and there were two coal-fired chimneys around the DJU sampling site. The central heating in the winter resulted in a lot of smoke and dust emissions, and brought more particulate pollutants into the air. The thermal inversion weather pattern was more frequent in winter than in other seasons, so that the pollutants in the air could not disperse very well.

There are two important ports close to the DMO sampling site, which are DLP and DNP. DLP is the most important and integrated foreign trade port in northeast China, and DNP is a modern deepwater oil port. It is well-known that an oil pipeline explosion occurred at DNP on 16 July, 2010, which was likely to affect the concentration and TEQ values of PCBs in the atmosphere of Dalian. **Table 2** shows that the concentration of the PCBs in the air increased at the two sampling sites in July, but the TEQ value of PCBs did not increase at the DJU sampling site, while the TEQ value of PCBs at the DMO sampling site increased slightly compared to that in June. The sample at the DJU sampling site in July was collected before the pipeline explosion (11–13 July, 2010), so that the concentration change of PCBs at DJU in July was not affected by the oil pipeline explosion event. The sample at the DMO sampling site in July was collected after the pipeline explosion (19–23 July, 2010), and although the concentration and TEQ value of PCBs in the air were increased at the DMO sampling site, the concentration variation trends at the two sampling sites were consistent in June, July and August. It can be considered that the oil pipeline explosion at DNP had some minor influence on the concentration and TEQ value of PCBs of the urban air in Dalian.

### 2.4 Correlation of the concentration of PCBs with meteorology parameters

PCBs exist in air as vapor and associated with suspended particles. Adsorption to aerosols depends on the vapor pressure of the compounds, the amount and the type of particulate matter present and the ambient temperature (Falconer and Bidleman, 1994). Correlation analysis of meteorological parameters with the concentrations of PCBs was conducted using SPSS packages. The Spearman rank correlation coefficients are listed in **Table 4**.

**Table 4** shows that the concentrations of  $\Sigma$ PCBs in the gas phase (PUF) had much higher negative (−0.635\*) and positive correlations (0.551) with the atmospheric pressure and ambient temperature respectively than the

**Table 4** Spearman rank correlation coefficients of PCBs concentration with meteorological parameters

	TSP	Pressure	Temperature	Relative humidity	Wind speed	ΣPCB (PUF)	ΣPCB (GFF)
TSP	1						
Pressure	0.382	1					
Temperature	−0.246	−0.860**	1				
Relative humidity	−0.269	−0.353	0.613*	1			
Wind speed	−0.030	−0.396	0.124	0.062	1		
ΣPCB (PUF)	−0.395	−0.635*	0.551	0.470	−0.016	1	
ΣPCB (GFF)	0.199	0.676*	−0.907**	−0.471	−0.032	−0.369	1

TSP: total suspended particle. \*\* Correlation is significant at the 0.01 level (2-tailed); \* correlation is significant at the 0.05 level (2-tailed).

other meteorological parameters. In contrast, those in the particulate phase (GFF) were correlated positively with atmospheric pressure (0.676\*), and negatively with temperature (−0.907\*\*). This means that the concentrations of PCBs in PUF decreased with increasing atmospheric pressure, and increased with increasing temperature, but those in GFF displayed a reverse trend. The influence of ambient temperature on the concentrations of PCBs in PUF was consistent with the results observed by Kim et al. (2011) in South Korea, Lohmann et al. (2000a) in England, and Bozlaker et al. (2008) in Izmir, Turkey. The concentration of PCBs had little correlation with TSP, relative humidity or wind speed. But roughly speaking, the concentration of PCBs in the gas phase was positively correlated with air humidity and negatively with TSP. However, the concentrations of PCBs in the particulate phase displayed just the reverse trend. The concentrations of PCBs in both phases all showed a few negative correlations with the wind speed.

Wania et al. (1998) suggested that high temperature de-

pendence indicates that the air concentration is controlled by revolatilization from the surfaces in the local surrounds of the sampling sites, whereas low temperature dependence indicates that the atmospheric transport of an air mass controls the air concentration. From that point of view, the volatilization of PCBs from local surfaces would dominate the urban air concentrations of PCBs in Dalian.

## 2.5 Relationship between SPMD samples and active samples

SPMD samples were sampled after they were deployed for 52 days, three months, half year and one year, separately.

**Table 5** shows the concentration of PCBs sampled by SPMDs for three months (22 Apr 2010–23 Jul 2010) and the average concentration of PCBs in three months (May, June, July) with active sampling. **Table 5** confirms the pronounced abundance of indicator PCBs in comparison to DL-PCBs and the dominant contributors to DL-PCBs were PCB118, PCB105 and PCB77 in both kinds of the samples.

**Table 5** Concentrations and WHO-TEQ of PCBs in SPMDs and active sampling samples

Compound	DJU			DMO		
	C <sub>SPMD</sub> (pg/SPMD)	C <sub>air-gas</sub> (pg/m <sup>3</sup> )	C <sub>air-particles</sub> (pg/m <sup>3</sup> )	C <sub>SPMD</sub> (pg/SPMD)	C <sub>air-gas</sub> (pg/m <sup>3</sup> )	C <sub>air-particles</sub> (pg/m <sup>3</sup> )
Indicator PCB						
PCB28	1306	4.38	0.17	1335	5.97	0.17
PCB52	1233	1.75	0.022	1283	2.84	0.025
PCB101	1711	2.47	0.052	1478	3.12	0.033
PCB138	880	0.30	0.032	606	0.40	0.034
PCB153	1029	0.31	0.028	722	0.48	0.028
PCB180	297	0.12	0.035	163	0.11	0.032
Non-ortho PCB						
PCB77	125	0.18	0.017	112	0.24	0.027
PCB81	26.9	0.050	0.003	21.8	0.091	0.004
PCB126	n.d.	0.022	0.005	7.7	0.030	0.008
PCB169	n.d.	0.007	0.005	n.d.	0.008	0.005
Mono-ortho PCB						
PCB105	383	0.40	0.048	374	0.80	0.060
PCB114	49.3	0.46	0.003	49.0	0.085	0.004
PCB118	1077	1.12	0.094	1027	2.36	0.11
PCB123	57.6	0.036	n.d.	50.6	0.033	n.d.
PCB156	75.6	0.91	0.033	50.5	0.18	0.034
PCB157	24.4	0.66	0.008	5.9	0.039	0.009
PCB167	34.0	0.081	0.014	27.0	0.12	0.015
PCB189	n.d.	0.010	0.007	n.d.	0.012	0.008
WHO-TEQ	0.23	3.51	0.54	0.98	3.61	0.87

n.d.: not detected.

**Table 6** Correlations of  $C_{\text{SPMD}}$  with  $C_{\text{air}}$ 

Sampling site	$C_{\text{air}} = aC_{\text{SPMD}} + b$	$a$	$b$	$r^2$
DJU	$C_{\text{SPMD}} - C_{\text{air-gas}}$	$1.0 \times 10^{-3}$	$6.6 \times 10^{-2}$	0.541
	$C_{\text{SPMD}} - C_{\text{air-particles}}$	$5.0 \times 10^{-5}$	$1.0 \times 10^{-2}$	0.421
	$C_{\text{SPMD}} - C_{\text{air-total}}$	$1.0 \times 10^{-3}$	$7.6 \times 10^{-2}$	0.543
DMO	$C_{\text{SPMD}} - C_{\text{air-gas}}$	$2.0 \times 10^{-3}$	$-1.4 \times 10^{-1}$	0.766
	$C_{\text{SPMD}} - C_{\text{air-particles}}$	$5.0 \times 10^{-5}$	$1.2 \times 10^{-2}$	0.417
	$C_{\text{SPMD}} - C_{\text{air-total}}$	$2.0 \times 10^{-3}$	$-1.3 \times 10^{-1}$	0.763

Correlation analysis of the concentration of PCBs sequestered by SPMDs ( $C_{\text{SPMD}}$ ) with the concentrations of PCBs in gas phase ( $C_{\text{air-gas}}$ ), particulate phase ( $C_{\text{air-particles}}$ ) and the air (gas phase + particulate phase,  $C_{\text{air-total}}$ ) was conducted. There were a certain number of linear correlations between  $C_{\text{SPMD}}$  and  $C_{\text{air-gas}}$ ,  $C_{\text{SPMD}}$  and  $C_{\text{air-total}}$  (Table 6). Moreover, the values of  $a$ ,  $b$  and  $r^2$  of the correlation analysis of  $C_{\text{SPMD}}$  versus  $C_{\text{air-gas}}$  and  $C_{\text{SPMD}}$  versus  $C_{\text{air-total}}$  were similar. This indicated that SPMD mainly sequestered gas phase PCBs.

### 3 Conclusions

The average concentration of PCBs in the gas phase was higher than that in the particulate phase in the urban air of Dalian. The dominant PCB components in each season were low and middle molecular weight PCBs. With increasing chlorination level, the concentration of the congeners of PCBs in the air decreased. Generally, absorption mechanisms contributed more to the gas-particulate partitioning process than adsorption. The concentration of PCBs in the gas phase decreased with the increase of atmospheric pressure, and increased with the increase of ambient temperature, but in the particulate phase a reverse trend was displayed. The main contributors to the  $\Sigma$ DL-PCBs concentration were PCB118, PCB105 and PCB77 and the dominant contributor to WHO-TEQ was PCB126 for active and SPMD passive samples. SPMD mainly sequestered gas phase PCBs.

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### Supporting materials

Supporting materials related to this article can be found in online version.

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## Supporting Information Available

The meteorology parameters and TSP values during sampling periods (**Table S1**); Concentrations and TEQ of  $\Sigma$ PCBs in the atmosphere in the cities around the world (**Table S2**) are listed.

**Table S1** Meteorology parameters and TSP values during sampling periods

Data	P (hpa)	Temp. (°C)	RH (%)	WS (m/sec)	TSP (ug/m <sup>3</sup> )
Nov. 2009	1013.9	1.0	75.3	3.7	96.7
Dec. 2009	1015.7	-4.5	54.5	3.4	128.4
Jan. 2010	1018.2	-6.2	68.3	2.5	64.9
Mar. 2010	1003.2	2.2	63.0	4.1	82.9
April 2010	1006.8	7.8	61.2	2.8	65.0
May 2010	1000.1	16.3	62.3	3.7	92.6
June 2010	994.7	20.7	69.0	3.9	51.3
July 2010	997.3	23.1	92.0	3.6	64.3
Aug. 2010	1001.8	25.0	85.5	3.1	103.6
Sept. 2010	1003.7	20.2	71.5	2.7	91.0
Oct. 2010	1007.9	13.3	78.8	3.9	70.4

**Table S2** Concentrations (pg/m<sup>3</sup>) and TEQ of  $\Sigma$ PCBs (fg-TEQ/m<sup>3</sup>) in the atmosphere in the cities around the world

Sampling site	Sample time	Category	$\Sigma$ PCBs	TEQ	Reference
Dalian, China	2009.11–2010.10	PUF+TSP	18.61–91.02	3.57–22.07	This study
Taiwan, China	1994	PUF+TSP	3480–7830(5910)		Chen et al., 1996
Guangzhou, China	2001, W	PUF+TSP	176.43		Li et al., 2004
Guangzhou, China	2005–2006, W	TSP	223	450–3000	Ren et al., 2010
Pearl River Delta, China	2001–2002, W	PUF+TSP	435.57–469.60		Li et al., 2007a
Shanghai, China	2005–2006, W	PM <sub>10</sub>	33.54–637.48	0–14.03	Li et al., 2007b
Beijing, China	2005–2006, W	TSP	5.52(ng/g)	2.27(pg/g)	Liu et al., 2007
Beijing, China	2008, W	PM <sub>2.5</sub>	7.2 16.2	13.58	Sun et al., 2009
Beijing, China	2007–2008, W	PUF+TSP	1295.49–1549.74		Li et al., 2009
Chicago, USA	1996–2003	PUF	100–9500(1400)		Sun et al., 2006
Chicago, USA	1995	PUF+TSP	1900 ± 1700		Tasdemir et al., 2004
Chicago, USA	1997–1999, S&A	PUF+TSP	2400–4100		Strandberg et al., 2001
Chicago, USA	2008, S	PUF+TSP	1130 ± 580	0.922	Persoon et al., 2010
Baltimore, USA	1997, S	PUF+TSP	760–2220		Brunciak et al., 2001
Milwaukee, USA	2001, S	PUF+TSP	1900 ± 780		Wethington et al., 2005
Pairs, France	1990	PUF+TSP	2000–6000		Granier et al., 1997
Landon, Manchester, Cardiff, England	1991–1992	PUF+TSP	112–3850		Hlasall et al., 1995
Athens, Greece	2000, S	PUF+TSP	348.6		Mandalakis et al., 2002
Seoul, South Korea	1999–2000, A&W	PUF+TSP	130.41 ± 62.57		Yeo et al., 2004
Bursa, Turkey	2004–2005	PUF+TSP	491.8 ± 189.4	4.409	Cindoruk et al., 2007
Yokohama, Japan	2002–2003	PUF+TSP	62–250	2–14	Kim et al., 2005

S: Summer, A: Autumn, W: Winter.

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