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Polychlorinated Biphenyls (PCBs) in PM_{10} Surrounding a Chemical Industrial Zone in Shanghai, China

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Abstract In order to gain comprehensive understanding of status, properties and sources of PCBs pollution at an industrial area in Shanghai, PM_{10} were collected during the period November 2004–September 2005. The results showed that the mean value of total PCBs in the industrial area was 2,017.22 pg m^{-3}. Three dioxin-like PCB congeners had a mean value of TEQ of 0.24 pg-TEQ m^{-3}. The concentrations of PCBs at all sites were higher in colder months than in warmer months. ΣPCB concentrations were correlated positively with SO_2, NO_2 and OCPs, while negatively with polycyclic aromatic hydrocarbons (PAHs), ambient temperature, rainfall and wind speed. It could be concluded that the area had been contaminated by PCBs from a local source.

Keywords PM_{10} · PCBs · Industry area · Seasonal character

Polychlorinated biphenyls (PCBs) are considered as significant environmental contaminants due to their ecotoxicological and human health risk (Loganathan and Kannan 1994; Toan et al. 2007; Merivirta et al. 2006). Facing the persistent risks posed by the manufacture and use of PCBs, several researches about environmental contamination from PCBs have focused on the industrial districts in Italy (Rossini et al. 2005), Spain (Schuhmacher et al. 2004), Russia (Milosevic-Djordjevic et al. 2004), and Republic of Azerbaijan (Swartz et al. 2003) and the USA (Kannan et al. 1997). While in China, many studies have focused on the aquatic ecosystems in eastern China (Schauer et al. 2002; Zhang et al. 2002; Yang et al. 2005), especially in estuarine areas such as those of the Pearl and Yangtze Rivers, Bohai Bay on the east coast of North China, and along other major rivers such as Minjiang, Jiulongjiang and Songhua (Xing et al. 2005). However, contamination status in terrestrial ecosystems especially in industrial zones was not studied yet. In fact, atmospheric pollution has become severe increasingly in China due to its rapid urbanization and industrialization. Especially in Shanghai a typical metropolis of China, which has several large industrial regions. In this study, we analysed the properties and sources of PCBs in PM_{10} surrounding a chemical industrial zone in Shanghai (Wujing industrial zone). The objectives of this study were to investigate seasonal variation of PCB concentrations in the area, to understand the behavior of PCBs in the atmosphere through gas/particle partitioning, and to identify the sources of PCBs in ambient air.

Materials and Methods

Wujing industrial zone (N31°00′, E121°25′) is situated in the southeast of Minghang District, which is in the center of Shanghai municipality with a population of 1.6 million. It is one of the important chemical industrial zones in Shanghai, with 50 years history, it lies on the upriver of the Huangpu River. The yearly average temperature is 15.5°C, and there are no major obstructions from east to south more than 1 km in the primary wind directions. This industrial area is based on coal as the fuel, and has already formed the industry chain with Coking & Chemical Plant. Coal-fired Power plant (CFP), Chlor-Alkali Chemical factory (CAC) and Coking &

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Chemical factory (CCF) in Wujing industrial area were selected for PM_{10} collection (Fig. 1). CFP has a capacity of 1.2 million kW, CFP site is located on the southeastern side of the factory and is about 500 m away from the smokestack. CAC is one of the largest chlor-alkali enterprises in China. It produces more than ten kinds of chemical products such as caustic soda, liquid chlorine, hydrochloric acid, sodium hydrosulfite, PVC, ferric sodium hypochlorite, triphosgene, and insecticide series, and so on. CAC site is located on the western side of the factory and is about 500 m away from its emission sources. CCF corporation is a comprehensive chemical company using coal as a major raw material and is the biggest manufacturer of city gas with a daily output 3.2 million m^3, half of the city's usage. CCF site is located on the northeastern side of the factory and is about 500 m away from its emission sources.

PM_{10} were collected every 2 months from November 2004 to September 2005, for four seasonal sampling periods: spring (March and May), summer (July), fall (September and November) and winter (January). Mean temperature during sampling in spring was 15°C, in summer was 29°C, in fall was 21°C, and in winter was 3.8°C. KB-1000 PM_{10} sampler (Laoshan Electronic Instrument Company, Qingdao, China) was set at each site for collecting ambient particles. Air inlets were 1.5 m above the ground, and the air flow was set at 1.01 m^3 min^{-1}. Glass fiber filter (200 × 250 mm^2) was baked at 450°C for 4 h to remove trace organics before using for sample collection. PM_{10} collection started at 9:30 a.m. and ended at 15:30 p.m. Sampling dates were selected according to the following meteorological aspects: (1) there was no rain over four consecutive days prior to sampling, (2) the atmospheric temperature was of seasonal characteristics. Before and after collection, filters were wrapped in baked aluminum foil. The mass of PM_{10} was determined by weighing the filters before and after exposure. Prior to weighing, the filters were placed in a constant temperature and humidity condition at 25°C and 50%, respectively, for 24 h. After weighing, the samples were wrapped in aluminum foil and stored at –4°C until analysis.

The analysis of the samples was performed according to the US EPA method 8000 series. Samples were analyzed for 33 PCBs congeners. Details of the analytical methods can be found elsewhere (Kim and Masunaga 2005). Prior to extraction, samples were fortified with ^{13}C-labeled PCB-209 (IUPAC number), which served as a surrogate for assessing the method recoveries for each sample. All samples were ultrasonic extracted for 3 min with hexane–acetone (1:1 volume). The extracts were decanted through separatory funnels containing filter paper and anhydrous sodium sulfate. The extraction was repeated three times. All extracts were combined and then centrifuged for 5 min at 1,500 rpm. Supernatant was concentrated by rotary evaporator to less than 10 mL, and then was solvent exchanged by adding 50 mL of *n*-hexane. A small volume of extracts was treated repetitively with concentrated sulfuric acid until a clear and colorless *n*-hexane extract was obtained. The hexane extract was cleaned up on a silica gel/anhydrous sodium sulfate column. One gram of anhydrous sodium sulfate was transferred over a layer of 3 g of silica gel (3% distilled water deactivated) packed in a glass column (25cm × 10mm i.d.) with Pyrex glass wool at bottom and a PTFE stopcock. PCB congeners were eluted with *n*-hexane (80 mL). And the extract volume was concentrated to 1 mL at room temperature under a gentle stream of ultrapure nitrogen. Analytical standards were the mixture of Aroclor 1254, Aroclor 1242 (Supelco Inc., Bellefonte, PA) and the surrogate PCB209 (AccuStandard Inc., New Haven, CT). All solvents were pesticide grade. Prior to their use, silica gel was baked at 400°C.

Quantification was performed on a GC (Agilent 6890N) equipped with a ^{63}Ni electron capture detector (GC-ECD)

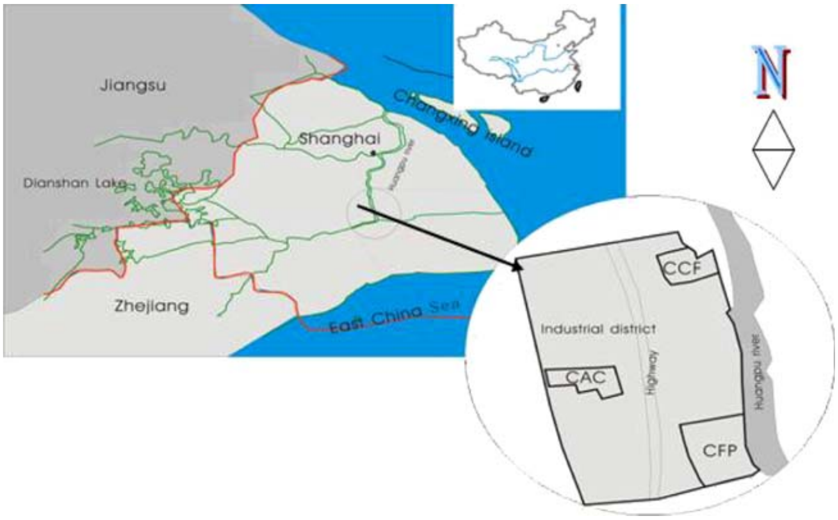

Fig. 1 Location map of the sampling station in Wujing industrial area, Shanghai, China

and a HP-5 capillary column (30.0m × 0.32mm × 0.25μm) using external standard solutions, and the following instrumental conditions: splitless injector at 225°C, detector at 300°C, initial oven temperature at 100°C, hold for 2 min, heating to 160°C at 15°C min^{-1}, then to 270°C at 5°C min^{-1}, with final 20-min hold. Oxygen-free nitrogen (99.999%) was used as carrier and make up gas. Qualitative analysis for PCB congeners was carried out on a GC-MS (Shimadzu QP2010) in EI mode monitoring selected ions and a CBP1 capillary column (25 m long, 0.22 mm i.d., 0.25 μm film thickness stationary phase) for confirmation of results.

About 10% of the samples were method blanks (MB), and a laboratory control sample (LCS) was included with each analytical batch, prepared and treated in an identical fashion to the samples. This also included the analysis of at least one matrix spike/matrix spike duplicate (MS/MSD) paired with each batch of up to 20 samples of the same matrix processed together. The method detection limits (MDL) was defined as the mean plus three times the signal/noise of the MB. The method had an overall recovery of 87.6%–130%. Reported concentrations were not corrected for the recovery.

Results and Discussion

The average concentrations of PM$_{10}$-bound PCBs (sum of the examined congeners) in ambient air are shown in Table 1. PCB congeners are identified by their International Union of Pure and Applied Chemistry (IUPAC) nomenclature. The ΣPCB values were based on analyses of the congeners 8, 9, 19, 15/18, 16, 33/53, 22/51, 45, 52, 49, 47, 44, 37/42, 64, 74, 70, 95, 91, 84, 101, 85, 110, 82, 118/149, 114/134, 141, 138, 158, 128, 174, 156/171, 172 and 180 (33 congeners). The concentrations of total PCBs ranged from 40.75 to1,869.04 pg m^{-3} with a mean value of 535.81 pg m^{-3} in CAC site, and from 90.89 to 3,742.18 pg m^{-3} with a mean value of 1,736.98 pg m^{-3} in CFP site. Total PCBs presented the highest level of concentrations at CCF site, from ND to 14,149.16 pg m^{-3} with a mean value of 3,778.86 pg m^{-3}. The data on ambient concentrations of PCBs reported in literature are not directly comparable due to the different numbers of congeners determined as well as to the different targets of surveys; many investigators studied on the distribution of PCBs between vapor and particle phases. However, in an attempt of a rough comparison, some data concerning the particle phase PCBs are given below. The concentrations of particle phase PCBs in various US cities ranged from 9 to 108 pg m^{-3}. Lower concentrations were found in Sturgeon Point, New York and Lake Michigan (8.8 and 10 pg m^{-3}, respectively) (Hoff et al. 1996; Simcik et al. 1997). Higher concentrations were found in Chicago and New York Harbor (116 and 87 pg m^{-3}, respectively) (Simcik et al. 1997; Brunciak et al. 2001). Relatively higher concentrations had been reported for Madrid, Spain (García-Alonso et al. 2002). High concentrations, although with a decreasing trend in the past, had been reported for Paris, France (Granier and Chevreuil 1991; Biterna and Voutsa 2005). High concentrations of particle phase PCBs had been reported for a rural area in southern Taiwan (the particle phase represents 56.4% of the mean total concentration of 2,500 pg m^{-3}, Chen et al. 1996), whereas

Table 1 PCBs average concentrations in PM$_{10}$ in 1 year (pg/m^3) at an industrial area in Shanghai, China

IUPAC numbers	CAC	CCF	CFP
9	ND	ND	2.86
8	ND	ND	235.62
19	3.38	ND	3.17
15/18	1.73	4.13	18.70
16	5.65	ND	14.0
33/53	0.33	ND	1.35
22/51	5.01	4.36	4.48
45	ND	ND	0.81
52	0.40	ND	0.98
49	3.71	ND	3.8
47	0.27	13.98	20.24
44	147.92	7.66	12.22
37/42	0.38	ND	0
64	1.86	0.56	2.61
74	0.07	0.25	0.25
70	62.63	ND	0
95	0.25	ND	0
91	1.82	80.34	0.96
84	ND	0.59	4.47
101	0.33	205.12	0
85	74.35	768.88	0
110	ND	481.19	3.75
82	0.05	0.14	0
118/149	131.84	0.5	6.74
114/134	93.04	402.67	5.56
141	ND	424.45	2.6
138	0.19	0.04	0
158	ND	ND	1,384.10
128	ND	262.02	1.71
174	ND	ND	3.62
156/171	0.62	885.60	0
172	ND	5.45	0
180	ND	230.97	2.40
ΣPCBs	535.81	3,778.86	1,736.98
WHO-TEQ	0.06	0.65	0.0035

ND: under detection limit

relatively low concentrations were found in rural atmosphere of South Korea (6.56 pg m^{-3}) (Yeo et al. 2003). To assess the extent of how the concentrations at the industrial area compare with other locations, congener-specific concentration data (arithmetic means) reported for other locations were compiled. The mean value of total PCBs in the industrial area was 2,017.22 pg m^{-3}, which was 17–67 times higher than that in NW Greece (30 pg m^{-3}, Biterna and Voutsa 2005), Yokohama, Japan (62 pg m^{-3}, Kim and Masunaga 2005), New York Harbor and Chicago. This comparison suggests that the PCB levels in the industrial area were rather high, It might suggest that the area has been contaminated or it is still being contaminated by PCBs from a local source.

The compositions of PCB homologues in PM_{10} are shown in Fig. 2. The ratios of tetra-CBs to the total PCB concentration were the largest, followed by Tri-CBs > Penta-CBs > Hexa-CBs at CAC site. The ratio of Penta-CBs was largest, followed by Tetra-CBs > Hepta-CBs > Tetra-CBs > Tri-CBs > Hexa-CBs at CCF site. At CFP site, the ratio had the trend: Hexa-CBs > Tri-CBs > Tetra-CBs > Di-CBs > Penta-CBs > Hepta-CBs. The ratio of the tetra-CBs in our study ranged from 17.3% to 38.2% of the total concentration. Brunciak et al. (2001) reported that the ratio of tri-CBs and tetra-CBs ranged from 70% to 90% in coastal New Jersey in 1997–1999. Halsall et al. (1995) reported that the ratio of tri- and tetra-CBs was 50% in urban ambient air in the UK. The PCB homologue pattern was different among samples (Fig. 2). This indicated that aerial PCB concentrations at the sampling site were influenced by different emission sources.

TEQ concentrations were calculated using World Health Organization Toxic Equivalent Factors (WHO-TEFs) (Van den Berg et al. 1998), based on 17 PCDD/DF and 9 PCB congeners. As for dioxin-like PCBs, the levels of mono-ortho PCBs were higher than those of the non-ortho PCBs. In our study, PCB-118 was the predominant dioxin-like congener, followed by PCB-114 and PCB-156 in terms of concentration. The sum of the TEQ concentrations of the three dioxin-like PCB congeners (PCB-118, –114, –156) at the three sites was used to calculate TEQs (Table 1). The six additional dioxin-like PCB congeners with TEFs were not included in our analyses. The concentrations of TEQs ranged from ND to 0.23 pg-TEQ m^{-3} with a mean value of 0.06 pg-TEQ m^{-3} at CAC site, and from ND to 0.0085 pg-TEQ m^{-3} with a mean value of 0.0035 pg-TEQ m^{-3} at CFP site. Total TEQ was the highest at CCF site, from ND to 2.58 pg-TEQ m^{-3} with a mean value of 0.65 pg-TEQ m^{-3}. The three dioxin-like PCB congeners with a mean value of TEQ in the industrial area was 0.24 pg-TEQ m^{-3}. At CCF site in spring, given a person (60 kg body weight) was supposed to inhale 15 m^3 of air every day, personal intakes of the three dioxin-like PCB TEQs from the air would be calculated to be 0.65 pg kg^{-1} day^{-1}, which would be close to the acceptable daily intake or tolerable daily intake (WHO-12 PCBs, 1 pg kg^{-1} day^{-1}) proposed by several national and international organizations. To reveal the toxic equivalency factors (TEFs) for PCBs for humans at the sites, all the other six PCBs with TEFs need to be investigated. Furthermore, chlor-alkali facilities have been shown to release dioxins and furans (Kannan et al. 1998) and their human exposure to dioxins and dioxin-like compound may be much higher than the guidelines suggested by various health organizations.

The seasonal distribution of particle-associated PCBs is controlled by a combination of emission factors, dispersion conditions and chemical mechanisms. The quantities and characteristics of PCBs emitted from industrial stacks depended on several factors: type of fuel, manufacturing process, air pollution control devices, etc. This balance will depend on the relative importance of degradation processes and emission sources. The seasonal variation of PCB concentration is shown in Fig. 3. In Shanghai, winter is from November through January while summer is from May to July. Strong monsoon wind and dry weather are characterized as winter seasons, whereas hot and humid climate with occasional showers and thunderstorms represent summer seasons. Many prior studies had shown the increase of PCB concentration with higher temperature (Oehme et al. 1996; Joung et al. 2006). However, the total PCB concentrations in summer and autumn in this study were not the highest levels when temperature was the highest point. For the investigation of the seasonal variation of PCB concentrations, the year was divided in two periods, cold (winter and spring) and warm (summer and autumn), according to regional meteorological considerations. Specifically, during warm weather, the prevailing winds were due to thermal circulations with stronger

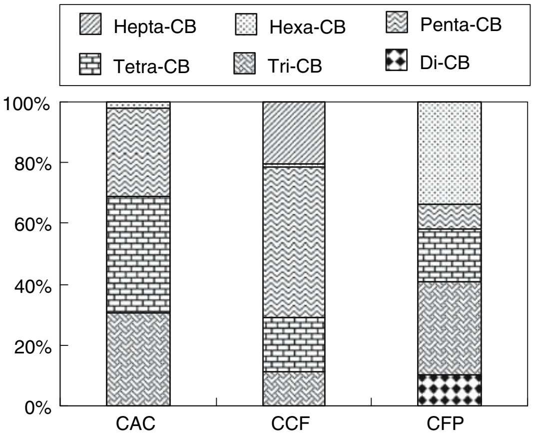

Fig. 2 Composition of PCB homologues in PM_{10} at an industrial site in Shanghai, China

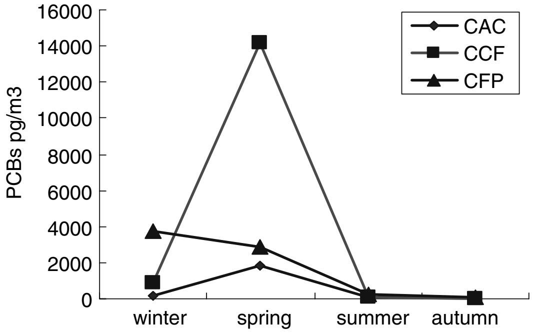

Fig. 3 Seasonal variations of PCBs in PM_{10} at an industrial site in Shanghai, China

mixing height and depth. The concentrations of PCBs at all sites were higher (and more variable) in cold weather than in warm weather (Fig. 3). The ratio of cold to warm total PCBs was 18.7 at CAC site, 147.7 at CCF site and 15.2 at CFP site, indicating significant seasonal variations in PCB concentrations. The low PCB concentrations in warm weather were mainly due to the high temperature, rainfall and the relatively clean air from the Huangpu River, which suggested that frequent storms with heavy rainfall in warm weather could remove PCBs effectively from the atmosphere. The substantial increase in PCB concentrations in cold weather in the industrial area was presumably caused by the light winds, the low effective photochemical degradation and the presence of fresh emissions. Higher concentrations of PCBs in atmospheric particles during cold period have also been observed by other investigators due to the lower ambient temperature as well as to the increase of suspended particles in the atmosphere (Bruncciak et al. 2001; Yeo et al. 2003).

The relationship of ΣPCB concentrations with air pollutants and meteorological parameters such as temperature, rainfall and wind speed is shown in Table 2. At all sites, ΣPCBs concentrations were significantly correlated with air pollutants, particularly the coal fire related SO_2. This is similar to that reported for PAHs in Korea (Moon et al. 2006). The combustion processes had been reported as the possible sources of PCBs and other POPs, which might be formed when a carbon source and chlorine were combusted together (Schoonenboom et al. 1995; Weber et al. 2001). The emissions from the combustion of different type of fuels (coal, wood, crude oil, leaded petrol, diesel), from various industrial activities and power generation processes were also found to contribute to PCB inventories (Harrad et al. 1994; Lohmann et al. 2000; Dyke et al. 2003). ΣPCBs concentrations were also found to correlate positively with NO_2 while negatively with PAHs. The relationship was possibly due to the secondary NO_2 formation, where time, dilution and environmental factors play an important role in dispersing the contaminants in the ambient atmosphere. Moreover, NO_2 had been implicated in the degradation of reactive PCBs on particles. Levels of PCBs and organochlorine pesticides (OCPs) were significantly correlated in all sites. The high correlation between PCBs and OCPs (mean $r > 0.3$; $p < 0.01$) indicates that they were likely to originate from the same source and that they represent a background pollution in this industry area. Assuming emission from local sources, negative correlation between ambient concentrations and wind speed should be considered, as a result of the accumulation of suspended particles during low wind periods in contrast to enhanced dilution and removal during stronger winds period. In the present study, strong negative correlations of ΣPCB concentrations with wind speed (significant at the 0.01 level) were found at CFP, where low wind speed prevailed (average wind speed 2.5 m s^{-1}) during the sampling period. A negative correlation of ΣPCB concentrations with wind speed (significant at the 0.05 level) was found at CAC and CCF (average wind speed 2.5 m s^{-1}). These correlations might suggest that PCBs were from local emission rather than regional transport. At all sites, ΣPCB concentrations were negatively correlated with ambient temperature and rainfall. This suggests that both meteorological parameters affect the vapor-to-particle partitioning of the semivolatile PCBs, and frequent storms with heavy rainfall could remove PCBs effectively from the atmosphere. The vapor pressures of these compounds have a well-defined temperature relationship, given by the Clausius–Clapeyron equation (Simcik et al. 1998; Xu et al. 2005).

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Table 2 Pearson correlation coefficients between ΣPCB concentrations and meteorological or other air pollution parameters in Shanghai, China

	SO_2	NO_2	PAHs	OCPs	Wind speed	Temperature	Rainfall
CAC	0.43*	0.40**	–0.36	0.98**	–0.39*	–0.50*	–0.57*
CCF	0.42*	0.40**	–0.39*	0.35*	–0.39*	–0.49*	–0.56*
CFP	0.75**	0.38*	–0.37	0.34*	–0.51**	–0.83**	–0.58*

* Significant at 0.05 level, ** significant at 0.01 level

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