Spatial character of polychlorinated biphenyls from soil and respirable particulate matter in Taiyuan, China

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ABSTRACT

As one of China's great metropolises, Taiyuan is affected by heavy chemical industry and manufacture of chemical products, and faces pollution from polychlorinated biphenyls (PCBs). Therefore, this study was conducted to determine the PCB concentrations in various environmental media in Taiyuan. We collected 15 soil samples, 34 respirable particulate matter (PM) samples (17 of PM_{2.5} and 17 of PM₁₀) from urban areas of Taiyuan, and measured a total of 144 PCB congeners (including some coeluting PCB congeners). The total PCB concentrations were $51\text{--}4.7 \times 10^3 \text{ pg g}^{-1}$ in soil, $27\text{--}1.4 \times 10^2 \text{ pg m}^{-3}$ in PM_{2.5} and $16\text{--}1.9 \times 10^2 \text{ pg m}^{-3}$ in PM₁₀. Of the PCB homologues, the dominant PCBs detected in the various media were all tri-CBs. Soil was relatively the most polluted media. Furthermore, principal-component analysis revealed that the major PCB source in Taiyuan may be associated with the main commercial PCB through long-range transmission. Toxic equivalency (TEQ) concentrations (based on ten dioxin-like PCBs) ranged from N.D. to $5.9 \times 10^{-3} \text{ pg-WHO TEQ g}^{-1}$ in soil, $2.0 \times 10^{-4}\text{--}3.4 \times 10^{-3} \text{ pg-WHO TEQ m}^{-3}$ and $1.0 \times 10^{-4}\text{--}1.2 \times 10^{-3} \text{ pg-WHO TEQ m}^{-3}$ in PM_{2.5} and PM₁₀, respectively. In previous studies, PCBs were not a severe component of contaminant in Taiyuan; however, this study suggested there is a potential threat of human exposure to PCBs for residents of Taiyuan.

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1. Introduction

Polychlorinated biphenyls (PCBs) were listed along with 11 other compounds as persistent organic pollutants (POPs) by the Stockholm Convention on 22 May 2001 (*The Secretariat of the Stockholm Convention and UNEPs Information Unit for Conventions*, 2005). PCBs are stable in the environment, can undergo long-range atmospheric transport, and can bioaccumulate through the food chain (UNECE, 1998; UNEP, 1998). PCBs are a class of 209 congeners that were widely used in a variety of electrical applications such as dielectric fluid in transformers and large capacitors, heat transfer fluids and hydraulic fluids. The carcinogenic and mutagenic properties of PCBs have been extensively studied (Breivik et al., 2002). Most research on environmental contamination from PCBs in China has focused on the eastern part of the country, especially estuarial areas such as those of the Pearl and Yangtze Rivers, Bohai Bay in northern China, and along other major rivers such as the Minjiang, Jiulongjiang, Yangtze, and Songhua Rivers (Xing et al., 2005a). However, the contamination status in terrestrial ecosystems, and especially in urban areas should not be ignored.

Soil and air are the important components of the terrestrial ecosystem. Soil is generally a primary sink and emission source for contaminants (Duarte-Davidson et al., 1997). Atmospheric particulates are important in contaminant transport over long distances in the terrestrial ecosystem (Hoff et al., 1998). The most significant exposure route of PCB is diet, and almost 95% of total daily intake (TDI) is taken up by dietary ingestion (*Japan Ministry of the Environment*, 2003). Direct human exposure via inhalation of outdoor air is not a significant exposure pathway, with respirable particulate matter (PM) the vector by which POPs enter the upper respiratory tract. Fine and ultra fine particles can reach lung alveoli, which is responsible for human exposure (Tasdemir et al., 2004; Kampa and Castanas, 2008). Since PCBs can easily accumulate on particles, contamination of soil and respirable PM have a direct impact on public health and increase the risk of human exposure via inhalation and ingestion (Tasdemir et al., 2004; Kampa and Castanas, 2008). Therefore it is important to monitor and improve understanding of various environmental media that influence breathing concentrations of such contaminants. Since the ratification of the Stockholm Convention (*The Secretariat of the Stockholm Convention and UNEPs Information Unit for Conventions*, 2005), it has become especially important to control PCBs in China. However, little is known about recent levels of PCBs in various environmental media of Chinese urban areas (Ren et al., 2007; Wang et al., 2008).

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As a metropolis, Taiyuan in Shanxi Province is probably one of the best-known industrial bases in China, with a long history of industrial activity, iron and steel industry, coal-fired power plants, and several coking and chemical plants around the urban area. Rapid economic growth and urbanization in recent years have sharply aggravated its pollution and raised a series of pollution problems. Thus it is necessary to immediately investigate the current contamination status of this key urban area in China.

Because of the paucity of data on PCB levels from various environmental media in Chinese cities (Meijer et al., 2003; Wang et al., 2008), we undertook this study to determine both the concentration and profile of PCBs in soil and respirable PM ($PM_{2.5}$ and PM_{10}) samples from Taiyuan urban areas. The main objectives were to establish the urban area's contamination status, explore the interaction of PCBs among these environmental media in the terrestrial ecosystem and identify possible sources of pollution.

2. Materials and methods

2.1. Samples

Taiyuan city occupies an area of 6956 km² with 180 km² classified as urban. The urban area is mainly in the eastern part of Taiyuan and has three million residents. China clay is the main representative soil type in Taiyuan. The climate is dominated by temperate semi-wet monsoon, with a mean annual temperature of -5.2°C during the sampling period.

Sampling sites were set to represent the area of Taiyuan. The sampling locations were chosen by a symmetrical grid (25 km² per grid), in an attempt to evenly distribute sampling over the region. Surface soil (0–10 cm) were obtained from 15 sites (site 1–15) in Taiyuan city, Shanxi province (Fig. 1), in January 2006. Surface soil (0–10 cm) was sampled in triplicate using a hand-held coring device from 15 sites (sites 1–15) in Taiyuan (Fig. 1), in Jan-

uary 2006. Each composite soil sample was 16 sub-samples from corresponding grids of sampling sites (Fig. 1). Overlying vegetation was removed prior to collection of the sample. The samples were freeze-dried, thoroughly mixed, sieved to 60-mesh, transferred to pre-cleaned amber glass and maintained at -18°C before analysis. The soil water content was determined gravimetrically after oven-drying individual composite soil samples at 105°C for 12 h.

Sampling was done in combination with the PCB soil study, using the same sampling sites. From the corresponding grid for soil samples (sites 1–17, Fig. 1) in Taiyuan, 34 respirable PM ($PM_{2.5}$ and PM_{10}) samples were taken in December 2006. To combine with the PCB soil study, the PM sampling was also done in winter. The PM samples were taken with a modified medium-volume TH-150 sampler with dual sampling module, which is in two main parts, filter holder and glass fiber filters, located on the top of the sampler (Wuhan Tianhong Instrument Factory, Wuhan, China). The overall average volume was approximately 24 m³ (average flow rate: 100 L min^{-1}) and each sample was a 4 h composite. There were 17 of $PM_{2.5}$ and 17 of PM_{10} samples taken. Before the experiment, the fiber filters were annealed for 4 h at 550°C to remove organic material and equilibrated in desiccators, weighed and put into acetone pre-washed aluminous envelopes. After sampling, the filters were removed from the inlet and folded in half and returned to the pre-clean aluminous envelopes and stored them at -18°C before analysis.

2.2. Chemicals

The standard solutions of PCB congeners were purchased from AccuStandard Inc. (New Haven, CT, USA). The five PCB calibration mixtures were diluted, which provided 144 congeners, with iso-octane to 20 ng mL^{-1} for each congener, which provided 144 congeners. Table 1 shows the 144 PCB congeners, which represent the commercial mixtures of PCBs most widely used, such as Aroclor

Fig. 1. Sampling sites of various environmental media in urban areas of Taiyuan.

Table 1

Acquisition data for GC–MS analysis of 144 PCB congeners including two selected ions for each homologue and relevant isotopic ratios.

PCB homologue	IUPAC number	Characteristic fragment ions MS-1	Characteristic fragment ions MS-2	Isotopic ratios
mono-CBs	1, 2, 3	188	190	3.13
di-CBs	4/10, 9/7, 6, 8/5, 14, 12/13, 15	222	224	1.56
tri-CBs	19, 30, 18, 17, 24/27, 16/32, 34, 29, 26, 25, 31, 28, 21, 33/20, 22	256	258	1.04
tetra-CBs	54, 53, 51, 45, 46, 69, 52/73, 49, 47/75/48, 44, 59/42, 71/41/64, 40, 67, 63, 74, 70, 66, 56/60, 81, 77	290	292	0.77
penta-CBs	104, 103, 100, 93, 95, 91, 92, 84, 90/101, 99, 119, 83, 97, 117/87/115, 85, 110, 82, 124, 107/109, 123, 118, 114, 122, 105	326	328	1.55
hexa-CBs	136, 154, 151, 135/144, 147, 149, 134, 165, 146, 153, 132, 141, 137, 130, 164/163/138, 158, 129, 128, 167, 156, 157	360	362	1.24
hepta-CBs	179, 176, 178, 175, 187, 183, 185, 174, 177, 171, 173, 172, 180/193, 191, 170/190, 189	394	396	1.05
octa-CBs	202, 201, 197, 200, 199, 203/196, 195, 194, 205	428	430	0.89
nona-CBs	208, 207, 206	462	464	0.77
dec-CB	209	496	498	0.69

1242, 1254, 1260, 1268 (World Health Organization (WHO), 1993). Decachlorobiphenyl (CB-209), used as a surrogate, was purchased from Supelco (Bellfonte, PA, USA). The working standards were prepared by diluting the above-mentioned standard solution with iso-octane. Standard reference material GBW08307 (reference soil for PCBs congeners analysis, including CB-28, 52, 77, 81, 101, 118, 114, 105, 138, 153, 167, 156, 157 and 180) was obtained from the National Research Center for Certified Reference Materials of China. The laboratory glassware was washed with detergents and dichromate acid cleaning solution, rinsed with tap water and distilled water, and then rinsed with methanol (analytical grade, Beijing Chemical Factory, China), dichloromethane (analytical grade, Beijing Chemical Factory, China) and acetone (analytical grade, Beijing Chemical Factory, China), respectively. The pre-clean procedure of each solvent was repeated six times using fresh solvent for the laboratory glassware prior to use. The other solvents used were pesticide grade (J.T. Baker, Phillipsburg, NJ, USA).

2.3. Extraction and cleanup

Five grams of each soil sample or each PM sample was ground with anhydrous sodium sulfate into a free-flowing powder. Each sample was extracted with 30 mL of hexane/dichloromethane (1:1, vol/vol) by ultrasonication for 4 min and then centrifuged at 3000g. This process was repeated three times and the extracts were combined. Before extraction, 100 μ L of CB-209 (100 ng mL⁻¹) was added in each sample as a surrogate standard and balanced at desiccator for 2 h. The concentrated extracts were evaporated to 1 mL in a Kuderna–Danish concentrator under a gentle N₂ stream for cleanup.

The concentrated extracts were cleaned by using a chromatography column (20 cm $\times$ 15-mm internal diameter) containing the following: 2 g of silver nitrate silica (10% concentrated silver nitrate, wt/wt), 1 g of activated silica gel, 3 g of basic silica gel, 1 g of activated silica gel, 4 g of acid silica gel (44% concentrated sulfuric acid, wt/wt), 4 g of acid silica gel (22% concentrated sulfuric acid, wt/wt), 1 g of activated silica gel and 2 g of anhydrous sodium sulfate. The PCB fraction was eluted with 100 mL of hexane. The elution was then evaporated with a rotary evaporator and then reduced to 50 μ L under a gentle N₂ stream for analysis.

2.4. Analysis

PCBs were measured with an Agilent 6890 series gas chromatograph (GC) coupled with an Agilent 5973 mass spectrometer (MS) using electron impact ionization source in SIM mode. In electron impact ionization mode, the MS source temperature was 230 °C, and the electron energy was 70 eV. The GC was equipped with a split-

splitless injector held constant at 270 °C. Gas chromatographic separation was performed on a DB-5MS capillary column (30 m $\times$ 0.25-mm internal diameter; 0.25- μ m film thickness). The GC column was maintained at 75 °C for 2 min and then ramped at 15 °C min⁻¹ to 150 °C, further ramped at 2.5 °C min⁻¹ to 280 °C, and held at this temperature for 15 min. The total run time was 64 min.

2.5. Quality control

All analytical procedures were monitored using strict quality assurance and control measures. Field blanks consisted of pre-clean soil and pre-clean glass fiber filters which were taken to the sampling site. A total of six field blanks were extracted and analyzed in the same way as samples. The target compounds were not detected when the field blank samples were analyzed. A solvent blank, a procedural blank and a standard mixture were run per 10 samples to check for contamination, peak identification and quantification. The target compounds were not detected in the solvent blanks and the procedural blanks. Duplicate samples in the laboratory were analyzed along with regular samples to determine repeatability and reproducibility, for additional quality-control assessment to ensure valid results. Instrument stability and relative response factor variance were determined by analyzing the calibration standard solutions in each sample batch.

PCBs were quantified using an external standard method. Three quality-control criteria were used to ensure correct identification of the target compounds: (1) GC retention times matched those of the standard compounds within 0.05 min; (2) the signal-to-noise ratio was greater than 3:1; and (3) each compound had two monitored ions. Isotopic ratios between quantitative and confirmation ions were within 15% of theoretical values. In the present study, some coeluting PCB congeners exist, and they were shown in Tables 1 and 2 (such as 4/10, 9/7, 8/5, 12/13 etc.). Coeluting PCB congeners were reported as individual results in Table 2. For example, PCB 9 and PCB 7 coeluted, so if the value of that coelution was 10, then the records for PCB 9 and PCB 7 will be shown with a single record containing the parameter name “PCB 9/7” and the quantitative value “10”. The limits of detection for PCBs were defined by a signal-to-noise ratio greater than three times the average baseline variation and were within 0.01–0.05 ng g⁻¹ (dry weight). The matrix spike recoveries of the 144 PCB congeners (including some coeluting PCB congeners) were within 70–110%, with 10% standard deviation. The recovery of CB-209 surrogate in all samples was within the range of 70–105%.

The standard reference material sample including 14 PCB congeners (GBW 08307, National Research Center for Certified Reference Materials, China) was analyzed to validate the analytical method used. The results were satisfactory, with a *z* score ≤ 1 for

Table 2Measured PCB concentrations in soil, PM_{2.5} and PM₁₀.

PCB congener (IUPAC No.) ^a	Soil (pg g ⁻¹ , n = 15)				PM _{2.5} (pg m ⁻³ , n = 17)				PM ₁₀ (pg m ⁻³ , n = 17)			
	Minimum	Maximum	Median	Standard deviation	Minimum	Maximum	Median	Standard deviation	Minimum	Maximum	Median	Standard deviation
1	N.D. ^b	63	N.D.	16	N.D.	1.7	N.D.	0.6	N.D.	1.9	1.1	0.8
2	N.D.	81	N.D.	26	N.D.	0.9	N.D.	0.2	N.D.	0.6	N.D.	0.2
3	N.D.	75	N.D.	24	N.D.	6.4	N.D.	2.0	N.D.	4.5	N.D.	1.1
4/10	N.D.	N.D.	N.D.	N.D.	N.D.	N.D.	N.D.	N.D.	N.D.	N.D.	N.D.	N.D.
9/7	N.D.	72	N.D.	19	N.D.	1.0	N.D.	0.2	N.D.	N.D.	N.D.	N.D.
6	N.D.	80	N.D.	28	N.D.	2.0	N.D.	0.8	N.D.	2.2	N.D.	0.8
8	N.D.	1.5 × 10 ²	N.D.	53	N.D.	6.7	N.D.	2.1	N.D.	12	N.D.	4.3
5	N.D.	1.1 × 10 ²	N.D.	37	N.D.	5.4	2.9	1.7	N.D.	7.4	3.1	2.3
14	N.D.	10	N.D.	2.7	N.D.	0.4	N.D.	0.1	N.D.	N.D.	N.D.	N.D.
12/13	N.D.	N.D.	N.D.	N.D.	N.D.	N.D.	N.D.	N.D.	N.D.	N.D.	N.D.	N.D.
15	N.D.	46	N.D.	13	N.D.	95	N.D.	23	N.D.	90	N.D.	24
19	N.D.	12	N.D.	5.5	N.D.	1.0	N.D.	0.4	N.D.	2.0	0.5	0.6
30	N.D.	3.4 × 10 ²	66	86	1.3	5.6	3.4	1.1	1.6	5.6	3.7	1.2
18	N.D.	4.5 × 10 ²	58	1.2 × 10 ²	N.D.	7.4	1.9	2.0	1.0	6.9	3.3	1.9
17	N.D.	18	N.D.	5.0	N.D.	0.5	N.D.	0.2	N.D.	1.3	N.D.	0.4
24/27	N.D.	3.2 × 10 ²	N.D.	83	N.D.	3.1	N.D.	1.2	N.D.	7.2	N.D.	2.1
16/32	N.D.	70	N.D.	27	N.D.	3.4	N.D.	1.1	N.D.	4.0	1.6	1.7
34	N.D.	17	N.D.	5.9	N.D.	1.2	N.D.	0.4	N.D.	0.6	N.D.	0.3
29	N.D.	11	N.D.	3.8	N.D.	0.7	N.D.	0.2	N.D.	0.8	N.D.	0.3
26/25	N.D.	9.5	N.D.	3.1	N.D.	0.8	0.3	0.3	N.D.	1.0	N.D.	0.4
31	N.D.	1.2 × 10 ²	23	38	N.D.	6.2	1.7	1.5	N.D.	4.2	1.9	1.1
28	N.D.	2.6 × 10 ²	N.D.	68	N.D.	2.4	N.D.	0.7	N.D.	2.4	N.D.	0.6
21	N.D.	19	N.D.	5.6	N.D.	12	N.D.	2.8	N.D.	12	N.D.	2.9
33/20	N.D.	34	N.D.	11	N.D.	N.D.	N.D.	N.D.	N.D.	2.7	N.D.	0.9
22	N.D.	30	N.D.	7.7	N.D.	0.8	N.D.	0.3	N.D.	0.7	N.D.	0.2
54	N.D.	17	N.D.	4.4	N.D.	0.6	N.D.	0.2	N.D.	N.D.	N.D.	N.D.
53	N.D.	22	N.D.	5.7	N.D.	N.D.	N.D.	N.D.	N.D.	0.9	N.D.	0.3
51	N.D.	81	N.D.	21	N.D.	5.1	1.0	1.7	N.D.	3.9	1.3	1.2
45	N.D.	24	N.D.	10	N.D.	3.9	N.D.	1.0	N.D.	0.9	N.D.	0.2
46	N.D.	24	N.D.	8.5	N.D.	0.8	N.D.	0.3	N.D.	2.1	N.D.	0.6
69	N.D.	16	N.D.	7.0	N.D.	0.9	N.D.	0.2	N.D.	1.0	N.D.	0.3
52/73	N.D.	1.1 × 10 ²	N.D.	30	N.D.	N.D.	N.D.	N.D.	N.D.	2.1	N.D.	0.7
49	N.D.	2.2 × 10 ²	N.D.	60	N.D.	1.1	0.5	0.4	N.D.	2.5	N.D.	0.7
47	N.D.	1.2 × 10 ²	N.D.	34	N.D.	N.D.	N.D.	N.D.	N.D.	1.0	N.D.	0.2
75/48	N.D.	1.1 × 10 ²	N.D.	35	N.D.	18	N.D.	6.0	N.D.	15	N.D.	3.6
44	N.D.	29	N.D.	10	N.D.	0.7	N.D.	0.2	N.D.	1.2	N.D.	0.4
59	N.D.	29	N.D.	10	N.D.	28	N.D.	6.7	N.D.	3.1	N.D.	0.9
42	N.D.	54	N.D.	16	N.D.	N.D.	N.D.	N.D.	N.D.	1.2	N.D.	0.4
71/41	N.D.	25	N.D.	9.1	N.D.	0.9	N.D.	0.3	N.D.	1.6	N.D.	0.5
64	N.D.	14	N.D.	4.6	N.D.	0.6	N.D.	0.3	N.D.	1.0	N.D.	0.3
40	N.D.	N.D.	N.D.	N.D.	N.D.	11	N.D.	2.8	N.D.	1.0	N.D.	0.2
67	N.D.	16	N.D.	5.3	N.D.	12	N.D.	3.0	N.D.	0.5	N.D.	0.2
63	N.D.	13	N.D.	3.3	N.D.	0.7	N.D.	0.3	N.D.	0.8	N.D.	0.3
74	N.D.	22	N.D.	7.3	N.D.	1.7	0.4	0.4	N.D.	1.5	N.D.	0.4
70	N.D.	3.9 × 10 ²	N.D.	1.0 × 10 ²	N.D.	1.8	0.4	0.5	N.D.	3.1	N.D.	0.8
66	N.D.	44	N.D.	12	N.D.	2.9	N.D.	0.7	N.D.	0.8	N.D.	0.3
56/60	N.D.	29	N.D.	9.9	N.D.	2.8	N.D.	0.7	N.D.	1.5	0.4	0.4
81	N.D.	11	N.D.	2.9	N.D.	10	0.5	2.5	N.D.	2.1	0.7	0.6
77	N.D.	20	N.D.	7.8	N.D.	3.9	0.8	1.0	N.D.	2.2	0.6	0.5
104	N.D.	N.D.	N.D.	N.D.	N.D.	N.D.	N.D.	N.D.	N.D.	0.6	N.D.	0.1
103	N.D.	N.D.	N.D.	N.D.	N.D.	N.D.	N.D.	N.D.	N.D.	0.8	N.D.	0.2
100	N.D.	N.D.	N.D.	N.D.	N.D.	0.7	N.D.	0.2	N.D.	6.7	N.D.	1.6
93	N.D.	N.D.	N.D.	N.D.	N.D.	N.D.	N.D.	N.D.	N.D.	0.9	N.D.	0.2
95	N.D.	N.D.	N.D.	N.D.	N.D.	1.0	N.D.	0.4	N.D.	0.9	N.D.	0.3
91	N.D.	15	N.D.	4.0	N.D.	0.7	N.D.	0.2	N.D.	1.6	N.D.	0.4
92/84	N.D.	24	N.D.	6.2	N.D.	N.D.	N.D.	N.D.	N.D.	1.6	N.D.	0.4
90	N.D.	1.3 × 10 ²	N.D.	35	N.D.	2.1	N.D.	0.6	N.D.	1.0	N.D.	0.3
101	N.D.	2.3 × 10 ²	N.D.	64	N.D.	1.4	N.D.	0.4	N.D.	1.3	N.D.	0.4
99	N.D.	22	N.D.	5.6	N.D.	2.5	N.D.	0.7	N.D.	0.9	N.D.	0.3
119	N.D.	12	N.D.	3.0	N.D.	0.8	N.D.	0.3	N.D.	1.0	N.D.	0.3
83	N.D.	N.D.	N.D.	N.D.	N.D.	1.3	N.D.	0.5	N.D.	1.8	N.D.	0.6
97	N.D.	29	N.D.	9.8	N.D.	1.0	N.D.	0.3	N.D.	1.2	N.D.	0.3
117	N.D.	17	N.D.	4.3	N.D.	3.6	N.D.	0.9	N.D.	0.9	N.D.	0.3
87	N.D.	N.D.	N.D.	N.D.	N.D.	0.8	N.D.	0.3	N.D.	0.8	N.D.	0.3
115	N.D.	81	N.D.	21	N.D.	0.8	N.D.	0.3	N.D.	0.8	N.D.	0.3
85	N.D.	27	N.D.	9.3	N.D.	1.1	N.D.	0.3	N.D.	1.3	N.D.	0.4
110/82	N.D.	6.2 × 10 ²	20	1.6 × 10 ²	N.D.	17	N.D.	4.1	N.D.	6.5	0.9	1.7
124	N.D.	45	N.D.	14	N.D.	1.1	N.D.	0.4	N.D.	2.5	N.D.	0.7
107	N.D.	16	N.D.	4.2	N.D.	1.2	N.D.	0.4	N.D.	1.0	N.D.	0.4
109/123	N.D.	15	N.D.	4.0	N.D.	2.3	N.D.	0.9	N.D.	1.7	N.D.	0.8
118	N.D.	N.D.	N.D.	N.D.	N.D.	1.3	N.D.	0.5	N.D.	10	0.6	2.5
114	N.D.	53	N.D.	14	N.D.	1.9	0.6	0.6	N.D.	2.1	0.7	0.6
122	N.D.	N.D.	N.D.	N.D.	N.D.	1.9	N.D.	0.6	N.D.	2.1	0.7	0.6

(continued on next page)

Table 2 (continued)

PCB congener (IUPAC No.) ^a	Soil (pg g ⁻¹ , n = 15)				PM _{2.5} (pg m ⁻³ , n = 17)				PM ₁₀ (pg m ⁻³ , n = 17)			
	Minimum	Maximum	Median	Standard deviation	Minimum	Maximum	Median	Standard deviation	Minimum	Maximum	Median	Standard deviation
105	N.D.	18	N.D.	4.7	N.D.	2.1	0.7	0.6	N.D.	1.8	0.6	0.6
136	N.D.	19	N.D.	4.8	N.D.	N.D.	N.D.	N.D.	N.D.	N.D.	N.D.	N.D.
154	N.D.	24	N.D.	6.2	N.D.	N.D.	N.D.	N.D.	N.D.	N.D.	N.D.	N.D.
151	N.D.	N.D.	N.D.	N.D.	N.D.	0.6	N.D.	0.2	N.D.	N.D.	N.D.	N.D.
135	N.D.	20	N.D.	5.0	N.D.	0.9	N.D.	0.2	N.D.	0.9	N.D.	0.2
144	N.D.	21	N.D.	6.3	N.D.	0.9	N.D.	0.2	N.D.	0.6	N.D.	0.2
147	N.D.	20	N.D.	5.0	N.D.	0.8	N.D.	0.2	N.D.	0.6	N.D.	0.2
149	N.D.	2.0 × 10 ²	14	57	N.D.	0.8	N.D.	0.2	N.D.	0.8	N.D.	0.2
134	N.D.	22	N.D.	5.7	N.D.	0.9	N.D.	0.2	N.D.	0.9	N.D.	0.2
165	N.D.	16	N.D.	4.2	N.D.	0.8	N.D.	0.2	N.D.	3.2	N.D.	0.8
146	N.D.	14	N.D.	4.5	N.D.	0.7	N.D.	0.2	N.D.	1.3	N.D.	0.3
153	N.D.	11	N.D.	4.5	N.D.	1.7	N.D.	0.4	N.D.	1.0	N.D.	0.2
132/141	N.D.	3.2 × 10 ²	N.D.	93	N.D.	2.0	N.D.	0.6	N.D.	1.9	N.D.	0.6
137	N.D.	26	N.D.	8.5	N.D.	N.D.	N.D.	N.D.	N.D.	N.D.	N.D.	N.D.
130	N.D.	32	N.D.	11	N.D.	0.9	N.D.	0.3	N.D.	1.3	N.D.	0.5
164	N.D.	34	N.D.	10	N.D.	0.8	N.D.	0.3	N.D.	1.1	N.D.	0.3
163/138	N.D.	3.3 × 10 ²	N.D.	87	N.D.	1.0	N.D.	0.3	N.D.	1.1	N.D.	0.3
158	N.D.	52	N.D.	14	N.D.	0.4	N.D.	0.1	N.D.	0.6	N.D.	0.2
129	N.D.	44	N.D.	14	N.D.	0.8	N.D.	0.3	N.D.	0.9	N.D.	0.3
128	N.D.	17	N.D.	5.6	N.D.	1.1	N.D.	0.4	N.D.	1.1	N.D.	0.4
167	N.D.	14	N.D.	5.5	N.D.	3.5	N.D.	0.9	N.D.	1.1	N.D.	0.3
156	N.D.	40	N.D.	11	N.D.	1.6	0.2	0.5	N.D.	1.4	0.6	0.4
157	N.D.	27	N.D.	10	N.D.	1.1	0.5	0.4	N.D.	0.9	0.4	0.3
179	N.D.	14	N.D.	4.3	N.D.	N.D.	N.D.	N.D.	N.D.	0.4	N.D.	0.1
176	N.D.	N.D.	N.D.	N.D.	N.D.	0.7	N.D.	0.2	N.D.	0.6	N.D.	0.2
178	N.D.	19	N.D.	4.9	N.D.	N.D.	N.D.	N.D.	N.D.	0.9	N.D.	0.3
175	N.D.	14	N.D.	3.6	N.D.	N.D.	N.D.	N.D.	N.D.	0.8	N.D.	0.3
187	N.D.	57	N.D.	15	N.D.	N.D.	N.D.	N.D.	N.D.	N.D.	N.D.	N.D.
183	N.D.	16	N.D.	5.2	N.D.	0.8	N.D.	0.2	N.D.	N.D.	N.D.	N.D.
185	N.D.	18	N.D.	7.0	N.D.	1.4	N.D.	0.3	N.D.	0.7	N.D.	0.2
174	N.D.	20	N.D.	6.4	N.D.	0.6	N.D.	0.2	N.D.	0.6	N.D.	0.2
177	N.D.	14	N.D.	3.5	N.D.	0.6	N.D.	0.2	N.D.	0.6	N.D.	0.1
171	N.D.	20	N.D.	6.7	N.D.	1.1	N.D.	0.3	N.D.	N.D.	N.D.	N.D.
173	N.D.	18	N.D.	6.1	N.D.	N.D.	N.D.	N.D.	N.D.	0.6	N.D.	0.2
172	N.D.	22	N.D.	7.1	N.D.	N.D.	N.D.	N.D.	N.D.	0.7	N.D.	0.2
180/193	N.D.	96	11	29	N.D.	1.5	N.D.	0.4	N.D.	0.8	N.D.	0.3
191	N.D.	12	N.D.	3.0	N.D.	0.5	N.D.	0.1	N.D.	0.9	N.D.	0.2
170/190	N.D.	1.1 × 10 ²	N.D.	29	N.D.	2.3	N.D.	0.6	N.D.	0.8	N.D.	0.3
189	N.D.	27	N.D.	9.3	N.D.	0.8	N.D.	0.3	N.D.	1.5	N.D.	0.4
202	N.D.	14	N.D.	3.7	N.D.	0.7	N.D.	0.2	N.D.	1.0	N.D.	0.4
201	N.D.	23	N.D.	8.6	N.D.	N.D.	N.D.	N.D.	N.D.	1.2	N.D.	0.3
197	N.D.	27	N.D.	9.5	N.D.	0.9	N.D.	0.3	N.D.	0.8	N.D.	0.3
200	N.D.	13	N.D.	3.3	N.D.	1.0	N.D.	0.2	N.D.	0.7	N.D.	0.2
199	N.D.	33	N.D.	11	N.D.	1.0	N.D.	0.3	N.D.	0.7	N.D.	0.2
203/196	N.D.	22	N.D.	8.1	N.D.	1.6	N.D.	0.4	N.D.	1.0	N.D.	0.4
195	N.D.	28	N.D.	10	N.D.	0.9	N.D.	0.3	N.D.	0.8	N.D.	0.3
194	N.D.	56	11.8	16	N.D.	1.3	N.D.	0.4	N.D.	1.1	N.D.	0.4
205	N.D.	55	N.D.	17	N.D.	1.0	N.D.	0.4	N.D.	1.1	N.D.	0.4
208	N.D.	32	N.D.	10	N.D.	1.1	N.D.	0.4	N.D.	0.9	N.D.	0.3
207	N.D.	20	N.D.	6.8	N.D.	N.D.	N.D.	N.D.	N.D.	1.4	N.D.	0.3
206	N.D.	41	N.D.	16	N.D.	1.7	N.D.	0.5	N.D.	1.4	N.D.	0.5

^a The number used to designate each congener by International Union of Pure and Applied Chemistry (IUPAC).

^b N.D. was calculated assuming non-detected measurements were equal to LOD.

all congeners (range of 0.02–0.64 for the PCBs; $p < 0.05$). Recovery of CB-209 surrogate was 85% with the GBW 08307 sample.

2.6. Data analysis

Statistical analysis was done using SPSS 13.0 for Windows (SPSS, Chicago, IL, USA). We used principal-component analysis (PCA), a multivariate statistical technique, to derive new components (PCs) as a linear combination of the original variables, where PCs attempt to preserve the statistical relationships of the original data. For this study, we investigated interspecies variations in PCB congeners using PCA. Initial variables considered were the concentrations of PCB homologues. We normalized values of PCBs to a percentage of the sum of all congeners. Varimax rotation in the PCA was used to facilitate interpretation.

3. Results and discussion

3.1. PCBs in soil

PCBs (144 congeners, including some coeluting PCB congeners) were identified in 15 urban soil samples (Table 2). The detection rates of PCBs in urban soils reached 100%, which indicates wide occurrence in Taiyuan (Fig. 2A). The total PCB concentrations (defined as the sum of 144 congeners) were 51–4.7 × 10³ pg g⁻¹ (median: 6.4 × 10² pg g⁻¹; dry weight; Table 3). These PCB levels were compared with soil concentrations of PCBs reported elsewhere (Ma et al., 2007; Ren et al., 2007; Fu et al., 2008; Wang et al., 2008). The median concentration in the present study was much lower than those obtained in global background soil (5.4 ng g⁻¹, dry weight) (Meijer et al., 2003) and was similar to

Fig. 2. Distribution of PCB homologues and Σ PCBs from various environmental media in Taiyuan (A: Soil; B: PM_{2.5} and PM₁₀).

those obtained in surface soil of China (median: 515 pg g⁻¹, dry weight) (Ren et al., 2007). In comparison with other areas of China, the levels were higher than those in suburban soil of Beijing (0.18 ng g⁻¹) (Chu et al., 1995); and lower than those in soil of Shenyang (6.4–15.2 ng g⁻¹), a major industrial city in north China (Jing et al., 1992), the surface soil of Dalian (2.8 ng g⁻¹, dry weight) (Wang et al., 2008), and soil of the chemical industrial zone in Shanghai (0.5–586.85 ng g⁻¹) (Ma et al., 2007). Compared to other countries, the levels were lower than those in surface soil of mainland Europe (97 ng g⁻¹) (Meijer et al., 2003) and Spain (4.4–4.7 ng g⁻¹) (Nadal et al., 2007), and were similar to those in the soil (0.6–5.1 ng g⁻¹) of the chemical/petrochemical industrial zone of Tarragona County (Nadal et al., 2008). According to the allowable level of 20 μ g kg⁻¹ for the total concentration of six PCB congeners approved by the Dutch List (VROM, 2000), the soil in our study generally had relatively low PCB contamination.

Of the PCB homologues, the dominant PCBs detected from soil in this study were tri-CBs (median: 28.8%), followed by tetra-CBs and di-CBs (medians: 17.2% and 13.1%, respectively; Fig. 2A). Higher-chlorinated congeners from soil were higher than those obtained from respirable PM in Taiyuan. This profile is reasonable since higher-chlorinated congeners are firmly bound to particles due to their low volatility (Chu et al., 1995).

3.2. PCBs in respirable PM

There were 144 PCBs (including some coeluting PCB congeners) identified in 34 respirable PM samples (17 of PM_{2.5} and 17 of PM₁₀, respectively; Table 2). The total PCB concentrations were 27–1.4 $\times 10^2$ pg m⁻³ (median: 46 pg m⁻³) in PM_{2.5}, and 16–1.9 $\times 10^2$ pg m⁻³ (median: 49 pg m⁻³) in PM₁₀ (Table 3). The median concentrations in PM_{2.5} and PM₁₀ in Taiyuan were similar. There are few previous studies on PM_{2.5} and PM₁₀ (Helm and Bidleman, 2005; Lammel et al., 2007; Mandalakis and Stephanou, 2007). Furthermore, atmospheric pollution directly influences respirable PM. Therefore, we compared the atmospheric concentrations of PCBs reported elsewhere (Cheng et al., 2007; Lammel et al., 2007; Mandalakis and Stephanou, 2007; Siddik Cindoruk and Tasdemir, 2007). In comparison with other areas of China, the median concentrations in the present study were lower than those from urban areas in the Yellow and East China Seas region (360 pg m⁻³ and 297–537 pg m⁻³, respectively) (Lammel et al., 2007), chemical industrial zone in Shanghai (2017.22 pg m⁻³) (Cheng et al., 2007). Compared to air from other countries, on the other hand, the levels were similar to those in the atmosphere of Melpitz (110 $\pm$ 80 pg m⁻³), a rural site in eastern Germany (Mandalakis and Stephanou, 2007), and were lower than those in the air from an urban area of Bursa, Turkey (491.8 $\pm$ 189.4 pg m⁻³) (Siddik Cindoruk and Tasdemir, 2007).

Of PCB homologues, the dominant PCBs in PM_{2.5} were similar to those in PM₁₀. The dominant PCBs detected from PM_{2.5} and PM₁₀ in this study were also tri-CBs, accounting for medians of 26.5% and 31.5%, respectively, followed by di-CBs and tetra-CBs (Fig. 2B). Higher-chlorinated congeners were detected at relatively low concentrations. This profile was expected since the lower volatility of higher-chlorinated congeners mean they become more firmly bound in heavily polluted areas (Chu et al., 1995). In other areas in China, tri-CBs were also the main PCB homologues in air samples (36.03–42.73%) of the Pearl River Delta (close to the City of Guangzhou, China) (Li et al., 2007). In urban air samples of Europe, the major PCB homologues were tetra-PCBs and tri-PCBs (Howell, 2007).

3.3. PCB spatial character

There were some correlations between soil and respirable PM (PM_{2.5} and PM₁₀). Concentration of Σ PCBs showed that soil was relatively the most polluted media, followed by PM_{2.5} and PM₁₀ (Table 3). According to each media, the differences in concentration might be neglected when accounting for errors in measurement, extraction and cleanup. These results indicated that in Taiyuan the PCB distributions in soil, PM_{2.5} and PM₁₀ were quite uniform. This may be due to the limited production of industrial PCBs and their relatively short application time in China.

In the present study, it was clear that the more chlorinated biphenyl congeners were higher in soil than in PM_{2.5} and PM₁₀, possibly due to their low volatility. Of PCB homologues, the tri-PCBs were dominant in all media in this study. Thus, soil was not only an important sink but also a key source that influenced respirable PM.

Toxic equivalency (TEQ) concentrations in soil and respirable PM (PM_{2.5} and PM₁₀) were calculated using World Health Organization Toxic Equivalent Factors (WHO-TEFs) (Van den Berg, 2006; Bhavsar et al., 2008). In the present study, TEQ concentrations based on 10 dioxin-like PCBs (PCB-77, PCB-81, PCB-105, PCB-114, PCB-118, PCB-123, PCB-156, PCB-157, PCB-167 and PCB-189). In the present study, PCB-81 was the predominant dioxin-like PCB, followed by PCB-77. The concentrations of TEQs ranged from N.D. to 5.9 $\times 10^{-3}$ pg-WHO TEQ g⁻¹ in soil, 2.0 $\times 10^{-4}$ –3.4 $\times 10^{-3}$ pg-WHO TEQ m⁻³ in PM_{2.5} and 1.0 $\times 10^{-4}$ –1.2 $\times 10^{-3}$ pg-WHO TEQ m⁻³ in PM₁₀, respectively (Table 3). Moreover, the most

Table 3

Total concentrations of PCB homologues and TEQs in each sample.

Sample	mono-CBs	di-CBs	tri-CBs	tetra-CBs	penta-CBs	hexa-CBs	hepta-CBs	octa-CBs	nona-CBs	ΣPCBs	TEQ
<i>Soil (the data of PCB concentrations are in pg g^{-1}; the data of TEQs are in pg-WHO TEQ g^{-1})</i>											
1	N.D. ^a	78	2.4×10^2	39	1.3×10^2	86	65	94	57	7.8×10^2	N.D.
2	63	N.D.	1.1×10^2	90	25	20	12	1.2×10^2	54	4.9×10^2	1.5×10^{-3}
3	N.D.	85	36	49	38	46	N.D.	17	21	2.9×10^2	2.0×10^{-3}
4	N.D.	88	1.7×10^2	68	4.7×10^2	4.5×10^2	36	N.D.	N.D.	1.3×10^3	1.8×10^{-3}
5	N.D.	N.D.	22	N.D.	N.D.	N.D.	N.D.	11	19	51	N.D.
6	2.0×10^2	1.7×10^2	2.7×10^2	60	1.2×10^2	13	11	N.D.	31	8.7×10^2	N.D.
7	N.D.	2.9×10^2	4.6×10^2	1.8×10^2	26	3.3×10^2	1.4×10^2	48	N.D.	1.5×10^2	3.0×10^{-3}
8	78	N.D.	1.3×10^3	7.6×10^2	1.1×10^3	9.8×10^2	3.6×10^2	88	24	4.7×10^3	3.1×10^{-3}
9	N.D.	1.0×10^2	84	1.1×10^2	95	2.7×10^2	1.6×10^2	1.2×10^2	36	9.8×10^2	6.0×10^{-4}
10	23	N.D.	4.0×10^2	2.0×10^2	76	1.3×10^2	12	67	N.D.	9.2×10^2	5.9×10^{-3}
11	N.D.	98	1.4×10^2	1.6×10^2	1.7×10^2	64	73	1.0×10^2	N.D.	8.1×10^2	1.3×10^{-3}
12	N.D.	N.D.	2.3×10^2	1.1×10^2	20	10	N.D.	N.D.	N.D.	3.7×10^2	2.1×10^{-3}
13	N.D.	1.4×10^2	1.4×10^2	2.6×10^2	N.D.	24	24	69	N.D.	6.5×10^2	N.D.
14	N.D.	1.5×10^2	5.1×10^2	4.0×10^2	2.1×10^2	2.0×10^2	1.0×10^2	43	34	1.6×10^3	1.1×10^{-3}
15	N.D.	N.D.	1.9×10^2	2.3×10^2	28	54	56	74	21	6.5×10^2	3.1×10^{-3}
<i>PM_{2.5} (the data of PCB concentrations are in pg m^{-3}; the data of TEQs are in pg-WHO TEQ m^{-3})</i>											
1	1.3	4.2	6.7	4.5	22	4.7	6.9	1.6	N.D.	52	5.0×10^{-4}
2	1.5	1.5	13	21	11	8.9	3.7	0.8	1.1	63	3.4×10^{-3}
3	0.4	3.0	12	69	8.7	7.9	0.7	N.D.	N.D.	1.0×10^2	9.0×10^{-4}
4	N.D.	13	8.1	22	11	2.7	N.D.	N.D.	1.7	58	5.0×10^{-4}
5	N.D.	14	5.4	3.0	N.D.	2.3	0.8	1.7	1.4	28	2.0×10^{-4}
6	7.3	4.0	12	20	5.2	3.5	N.D.	2.9	N.D.	55	1.0×10^{-4}
7	3.0	12	4.2	6.9	N.D.	1.4	N.D.	N.D.	N.D.	27	3.0×10^{-4}
8	0.6	5.1	9.4	7.6	6.5	2.0	1.1	N.D.	N.D.	32	2.0×10^{-4}
9	2.7	1.0×10^2	23	4.1	7.3	1.2	N.D.	1.4	N.D.	1.4×10^2	2.0×10^{-4}
10	4.5	14	9.7	6.6	7.7	1.8	0.6	0.6	N.D.	46	2.0×10^{-4}
11	1.1	8.7	13	25	1.2	0.6	N.D.	N.D.	N.D.	50	4.0×10^{-4}
12	2.4	2.0	11	7.5	1.6	2.0	N.D.	1.5	N.D.	28	3.0×10^{-4}
13	1.7	7.4	20	5.7	4.1	2.0	0.8	1.0	N.D.	42	2.0×10^{-4}
14	N.D.	14	11	6.0	13	5.4	0.6	0.6	N.D.	50	3.0×10^{-4}
15	N.D.	6.0	15	7.6	4.9	3.1	N.D.	1.7	1.0	39	2.0×10^{-4}
16	N.D.	11	13	5.3	4.0	2.8	N.D.	2.3	N.D.	38	2.0×10^{-4}
17	1.3	5.2	13	5.5	3.1	0.5	N.D.	0.6	N.D.	29	3.0×10^{-4}
<i>PM₁₀ (the data of PCB concentrations are in pg m^{-3}; the data of TEQs are in pg-WHO TEQ m^{-3})</i>											
1	1.9	9.3	11	5.4	16	9.3	6.5	0.6	N.D.	60	7.0×10^{-4}
2	N.D.	4.3	13	5.2	13	2.0	2.6	2.2	2.3	45	2.0×10^{-4}
3	N.D.	4.4	8.2	7.3	24	6.8	1.6	1.5	N.D.	53	8.0×10^{-4}
4	5.8	6.6	12	5.0	4.1	0.7	1.2	0.9	1.4	38	3.0×10^{-4}
5	N.D.	3.3	5.8	4.0	2.9	0.5	N.D.	N.D.	N.D.	16	1.0×10^{-4}
6	0.6	3.7	6.3	9.3	3.8	6.3	1.1	3.4	N.D.	35	5.0×10^{-4}
7	1.3	54	16	10	4.1	2.3	N.D.	1.1	N.D.	88	5.0×10^{-4}
8	N.D.	18	18	5.7	6.2	2.0	1.3	0.6	0.9	53	2.0×10^{-4}
9	1.7	N.D.	22	11	13	1.6	N.D.	N.D.	N.D.	49	4.0×10^{-4}
10	2.3	7.2	8.6	6.2	5.4	2.7	0.5	2.7	N.D.	36	6.0×10^{-4}
11	1.1	5.8	14	5.0	3.7	3.6	0.7	N.D.	N.D.	34	4.0×10^{-4}
12	N.D.	13	18	24	4.7	3.9	N.D.	4.5	N.D.	68	5.0×10^{-4}
13	1.8	1.1×10^2	40	13	26	2.1	N.D.	2.7	N.D.	1.9×10^2	1.2×10^{-3}
14	1.8	26	19	15	5.1	1.9	N.D.	N.D.	N.D.	68	2.0×10^{-4}
15	3.2	7.4	17	6.6	7.0	2.9	N.D.	1.3	0.7	46	2.0×10^{-4}
16	1.1	10	18	5.5	1.7	4.1	0.9	3.3	N.D.	44	2.0×10^{-4}
17	1.7	18	17	4.6	4.7	2.8	0.8	1.2	1.4	53	3.0×10^{-4}

^a N.D. was calculated assuming non-detected measurements were equal to LOD.

significant uptake route of dioxin-like PCBs is the diet (Japan Ministry of the Environment, 2003), with almost 95% of TDI via food contamination. Inhalation intake doses have been reported to contribute 2.61% to the TDI (Japan Ministry of the Environment, 2003). Due to lack of data about dioxin-like PCB diet exposure, we cannot calculate total daily dioxin-like PCB intake doses of Taiyuan citizens.

3.4. Source analysis

To evaluate similarities or differences between the PCB homologue patterns of various media samples, we performed PCA on data normalized to percentage of ΣPCBs. PCs were determined by an eigenvalue >1. By processing, two extracted PCs explained 70.3% of the data variance. A loading plot and score plot were obtained after varimax rotation. Component 1 accounted for 54.6% of

the total variance and was characterized by higher-chlorinated homologues, whereas component 2 accounted for 15.7% and was characterized by lower-chlorinated homologues. The score plot (Fig. 3) indicated that the compositions of PCBs in the various media samples were similar, possibly originating from the same source. The PCBs found in Taiyuan could therefore be classified into one main group that was relatively highly contaminated by tri-CBs. There was also a clear clustering of samples, showing that tri-CBs were the dominant contaminant in the soil, PM_{2.5} and PM₁₀.

In China, approximately 10000 t of PCBs were produced in 1965–1974, with World Health Organization (WHO), 1993. Polychlorinated biphenyls and terphenyls (2nd ed.), environmental health criteria 140. Geneva. Nine thousand tonnes of trichlorobiphenyl (the main PCB) and 1000 t of pentachlorobiphenyl (the #2 PCB) (Xing et al., 2005b); the former contained 42% chlorine, similar to Aroclor 1242, and the latter 53% chlorine, similar to

Fig. 3. Two-dimensional principal-component score plot obtained from the data correlation matrix of 15 soil samples, 17 samples of PM_{2.5} and 17 samples of PM₁₀ samples in Taiyuan.

Aroclor 1254 (Jiang et al., 1997). The major homologues produced and used in China was tri-CBs, the major homologue profile of global PCB products was also composed of tri-CBs, and the composition of tri-CBs (40.4% of total) was higher in Chinese products than in globally (25.2% of total) (Breivik et al., 2002). Therefore, tri-CBs should be the dominant PCBs in soil, PM_{2.5} and PM₁₀ of Taiyuan. All these samples as well as the major commercial PCB were characterized by a higher proportion of tri-CBs in the PCB profiles, which indicates that the sources of PCBs for these sampling locations may be associated with the main commercial PCB through long-range transmission.

4. Conclusions

The present work investigated the composition, distribution and characterization of PCBs in soil and respirable PM. The results indicate that the sources of PCBs for these sampling locations may be associated with the main commercial PCB through long-range transmission. Although the PCB contamination in various environmental media in Taiyuan was low, there was a potential threat to human health. All the findings indicate that attention should be paid to PCB contamination of the urban environment to maintain human health and environmental safety in China.

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