Polychlorinated biphenyl residues in sandstorm depositions in Beijing, China

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ABSTRACT

Sandstorms, which distribute many particles, are a special atmospheric occurrence and are frequent in northern China. We conducted this study to determine, for the first time, the concentration of polychlorinated biphenyls (PCBs) in sandstorm depositions. We collected 13 samples from urban areas of Beijing, and we measured a total of 144 PCB congeners. Thirteen samples all contained PCB residues. The total PCB concentration ranged from 1.6 to 15.6 ng g⁻¹ (median, 4.8 ng g⁻¹, dry weight), with trichlorinated biphenyls as the predominant homologue (>50.4%). Furthermore, we observed increasing PCB contamination from northwest to east Beijing. We later explored possible factors affecting contamination of the sandstorm depositions, which revealed a significant correlation between Σ PCBs and the minimum particle size of the sandstorm deposition samples. Principal-component analysis revealed that the major source of PCBs in Beijing may be potentially associated with the number-one commercial PCB through the long-range transmission. In previous results, PCBs were not a severe component of contamination in sandstorm depositions of Beijing. However, this study suggested that sandstorm deposition may be a potential source of exposure to PCBs for the residents of Beijing, China.

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1. Introduction

Polychlorinated biphenyls (PCBs), listed along with 11 other compounds as persistent organic pollutants by the Stockholm Convention on May 22, 2001, are stable in the environment, can undergo long-range atmospheric transport, and can bioaccumulate through the food chain (UNECE, 1998; UNEP, 1998; The Secretariat of the Stockholm Convention and UNEP's Information Unit for Conventions, 2005). PCBs are a class of 209 congeners that were widely used in variety of electrical applications such as the dielectric fluid in transformers and large capacitors, heat-transfer fluids, and hydraulic fluids. Therefore, PCBs have been extensively studied with regard to their carcinogenic and mutagenic properties (IARC, 1987, 1997). Research results published so far have focused on central and western Europe, North America, and Japan. There is particularly a shortage of data from China and Russia (Meijer et al., 2003).

Long-range atmospheric transport and deposition of PCBs are important factors to consider for controlling the regional and global distribution of these pollutants and other persistent organic pollutants as well as for assessing their effect on ecosystems. Furthermore, sandstorms, which distribute many particles,

are a special atmospheric occurrence and are frequent in northern China. For example, northern China experienced 11 sandstorms in 2006, one of which covered approximately one-eighth of China and deposited about 330000 tons of sand and dust in Beijing (Han et al., 2007; Lin and Chen, 2007). The regional characteristics of sand and dust sources are mainly arid or semiarid regions covered by deserts or land undergoing desertification in northern China (Wang et al., 2006). Wind-blown sand and floating dust not only remain in the areas where sandstorms occur but also extend to neighboring areas. The wind-blown sand extends either northeastward or southeastward, but floating dust extends mainly southeastward to low-latitude regions such as the East China Plain (Wang et al., 2003). Beijing, the capital of China, is one of the main cities affected by sandstorms and, with more than 12 million people, is one of the most densely populated cities in the world. The sources of human exposure to PCBs remain poorly understood. Natural and anthropogenic organic chemicals are transported within the atmosphere, while deposition events are important in helping to remove them (Scheringer et al., 2004). Because PCBs can easily accumulate in particles (Tasdemir et al., 2004), contamination of sandstorm deposition directly affects public health increasing the risk of human exposure by means of inhalation, ingestion or direct skin contact. We therefore need more information about the contamination status of PCBs in sandstorm depositions to better understand their environmental behavior and source–sink relationships.

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Because of the paucity of data on levels of PCBs in environmental media in China, we undertook this study to determine both the concentration and profile of PCBs in sandstorm deposition samples obtained from urban areas of Beijing. The main objectives of this study was to identify the possible sources of pollution in the sandstorm depositions of Beijing.

2. Materials and methods

2.1. Samples

A serious sandstorm developed from the Mongolian cyclone and blew over Beijing on April 16, 2006 (Han et al., 2007; Lin and Chen, 2007). The climate in Beijing then was dominated by temperate semiwet monsoon conditions, with a mean temperature of 14 °C during the sampling period. We obtained sandstorm deposition samples from 13 urban sites in Beijing, China (Fig. 1) on April 17, 2006. The total sampling region area was 750 km². We collected all samples from a platform 0.5–1.0 m in height and 3 m² in area during daytime (between 8:00 a.m. and 6:00 p.m. [10 h day⁻¹]) with no precipitation. The collected samples were composites of three subsamples obtained at each sampling site. We then transferred the gathered samples to precleaned amber glass bottles and stored them at 4 °C. Usually, we carried out extraction within 72 h. We collected and analyzed 13 samples for PCBs.

We analyzed the particle size distributions of the sandstorm deposition samples by using a Laser Particle Size Analyzer (Masterizer 2000; Malvern Co., United Kingdom).

2.2. Chemicals

We purchased the standard solutions of PCB congeners from AccuStandard Inc. (New Haven, CT, USA). We diluted the five PCB calibration mixtures, which provided 144 congeners, with *iso*-octane to 20 ng ml⁻¹ for each congener. Table 1 shows the 144 PCB congeners. Decachlorobiphenyl (CB-209) as a surrogate was purchased from Supelco (Bellfonte, PA, USA). Standard reference material National Institute of Standards and Technology (NIST) 2585 (reference house dust for organic contaminant analysis) was

obtained from NIST, GM, USA. All solvents used were of pesticide grade (J.T. Baker, Phillipsburg, NJ, USA).

2.3. Extraction and cleanup

We weight 5 g of each sandstorm deposition sample and then ground each one with anhydrous sodium sulfate into a free-flowing powder. We extracted the samples with 30 ml of hexane/dichloromethane (1:1, vol/vol) by ultrasonication for 4 min and then centrifuged the samples at 3000g. We repeated this process three times. Before extraction, we added 10 ng of CB-209 as a surrogate standard. We evaporated the concentrated extracts to 1 ml in the Kuderna–Danish concentrator under a gentle N₂ stream for cleanup.

We cleaned the concentrated extracts by using a chromatography column (20 cm × 15 mm, internal diameter) containing the following: 2 g of silver nitrate silica (10%, 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. We eluted the PCBs fraction with 100 ml of hexane. We then evaporated the elution with a rotary evaporator and then reduced it to 50 µl under a gentle N₂ stream for analysis.

2.4. Analysis

We analyzed PCBs 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 30-m DB-5MS capillary column (30 m × 0.25 mm, internal diameter; 0.25-µm film thickness). We maintained the GC column at 75 °C for 2 min and then ramped at 15 °C/min to 150 °C, further ramped at 2.5 °C/min to 280 °C, and held at this temperature for 15 min. The total run time was 64 min.

Fig. 1. Sampling sites of sandstorm deposition in urban areas of Beijing.

Table 1
Selected 144 PCB congeners

PCB homologue	IUPAC number
Mono-CBs	1, 2, 3
Di-CBs	4/10, 9/7, 6, 8/5, 14, 12, 13, 15
Tri-CBs	19, 30, 18, 17, 24/27, 16/32, 34, 29, 26, 25, 31, 28, 21, 33/20, 22
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
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
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
Hepta-CBs	179, 176, 178, 175, 187, 183, 185, 174, 177, 171, 173, 172, 180, 193, 191, 170, 190, 189
Octa-CBs	202, 201, 197, 200, 199, 203/196, 195, 194, 205
Nona-CBs	208, 207, 206
Dec-CB	209

2.5. Quality control

We ran a laboratory method control group to demonstrate the lack of interference and cross-contamination. We also ran a procedural blank in parallel with every set of six samples to further check for interference and cross-contamination. We analyzed duplicate samples in the laboratory along with the regular samples, for additional quality-control assessment to ensure valid results. We determined instrument stability and relative response factor variance by analyzing the calibration standard solutions in each sample batch.

We quantified PCBs by using an external standard method. We used three quality-control criteria 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; (3) each compound had two monitored ions. Isotopic ratios between quantitative and confirmation ions were within 15% of theoretical values. 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 the range of 0.01–0.05 ng g⁻¹ (dry weight). The matrix spike recoveries of the 144 PCB congeners were within the range of 70–110%, with a standard deviation of 10%. The recovery of CB-209 surrogate in all samples was within the range of 75–115%. The results met the acceptance criteria specified in the United States Environmental Protection Agency method 1668A (i.e., 25–150%) (Environmental Protection Agency, 1999).

We analyzed the standard reference material sample (NIST-2585) to validate the analytical method used. The results were satisfactory, with a *z* score ≤ 1 for all congeners (range, 0.06–0.84 for the PCBs; *p* < 0.05). Recovery of CB-209 surrogate was 85% with the

NIST 2585 sample, which also met the acceptance criteria specified in the US-EPA method 1668A.

2.6. Data analysis

The spatial distribution of PCBs was determined by Surfer 8.01 (Golden Software, Golden, CO, USA).

We carried out statistical analysis by using SPSS 13.0 for Windows (SPSS, Chicago, IL, USA). We used principal-component analysis (PCA), a multivariate statistical technique, to derive new components (principal components [PCs]) as a linear combination of the original variables, while PCs attempt to preserve the statistical relationships present in the original data. For this study, we investigated interspecies variations in the PCB congeners by using PCA. Initial variables considered were the concentration values of the PCB homologues. We normalized values of PCBs to a percentage of the sum of all congeners. To facilitate interpretation of results, we used rotation in the PCA with the varimax method.

3. Results and discussion

3.1. PCB composition

We identified PCBs in 13 sandstorm deposition samples. The total PCB concentrations (defined as the sum of 144 congeners) ranged from 1.6 to 15.6 ng g⁻¹ (median 4.8 ng g⁻¹, dry weight) (Table 2). There are few previous studies on sandstorm deposition. Furthermore, sandstorm deposition directly influences surface soil. Therefore, we compared the soil concentrations of PCBs reported elsewhere in the literature. The following areas were all affected by various degrees of sandstorm activity (Wang et al., 2003; Qiu et al., 2006). The median concentration in our study was similar to that of the soil of Beijing (3.1 ng g⁻¹) (Liu et al., 2006). In comparison with other areas surrounding Beijing, the levels were similar to those in agricultural surface soils of Shenyang (6.4–15.0 ng g⁻¹) (Jing et al., 1992), surface soils of Qingdao (3.1–15.0 ng g⁻¹) (Geng et al., 2006) and agricultural soils of Southern Jiangsu province (4.13 ng g⁻¹) (Zhang et al., 2007). This result indicated that the distribution of PCBs in sandstorm depositions in Beijing and in soils in Beijing and the surrounding region is much more uniform, possibly because of long-range atmospheric transportation (Meng et al., 2000). Furthermore, sandstorms might play a specific role in the long-range transmission. Because so far no threshold values of PCBs have been defined for soils or sandstorm depositions in China, we used as a reference of contamination the target value of the Dutch List that was formulated to distinguish between clean and polluted soils in The Netherlands (VROM, 2000). Compared with the allowable level of 20 μg kg⁻¹ for the total concentration of

Table 2
Total concentration of PCB homologues in each sample (data units are in nanograms per gram; ND, not detected)

Sample No.	Mono-CBs	Di-CBs	Tri-CBs	Tetra-CBs	Penta-CBs	Hexa-CBs	Hepta-CBs	Octa-CBs	Nona-CBs	ΣPCBs
1	N.D.	0.2	1.5	0.9	0.1	0.1	0.1	0.1	N.D.	2.9
2	N.D.	0.4	4.5	0.5	0.2	0.2	N.D.	N.D.	N.D.	5.8
3	N.D.	0.3	3.8	0.5	0.2	0.2	N.D.	N.D.	N.D.	5.1
4	N.D.	N.D.	2.8	0.7	0.4	0.2	N.D.	N.D.	N.D.	4.1
5	N.D.	1.4	10.7	2.5	0.6	0.3	0.1	N.D.	N.D.	15.6
6	N.D.	0.5	6.4	1.1	N.D.	0.1	0.1	N.D.	N.D.	8.2
7	N.D.	0.1	2.7	0.2	N.D.	0.2	0.1	N.D.	N.D.	3.4
8	N.D.	0.1	1.1	0.3	N.D.	0.1	N.D.	N.D.	N.D.	1.6
9	N.D.	0.1	2.5	N.D.	0.5	0.2	0.1	N.D.	N.D.	3.5
10	N.D.	N.D.	9.1	1.6	0.4	0.3	0.1	0.1	N.D.	11.4
11	N.D.	N.D.	4.5	0.2	N.D.	0.1	N.D.	N.D.	N.D.	4.8
12	N.D.	N.D.	1.9	0.3	0.5	0.2	N.D.	N.D.	N.D.	2.9
13	N.D.	0.5	8.7	1.7	0.6	0.5	0.1	N.D.	N.D.	12.1

six PCB congeners approved by the Dutch List, the contamination of sandstorm depositions in our study had relatively slight PCB contamination levels on average.

3.2. PCB profiles

In China, approximately 10000 tons of PCBs were produced from 1965 to 1974, with 9000 tons as trichlorobiphenyl (the #1 PCB) and 1000 tons as pentachlorobiphenyl (the #2 PCB) (Xing et al., 2005). The #1 PCB contained 42% chlorine, which was similar to Aroclor 1242, and the #2 PCB contained 53% chlorine, similar to Aroclor 1254 (Jiang et al., 1997). The major homologue produced and used in China was tri-CBs, the major homologue profile of the global PCB product was also tri-CBs, and the composition of tri-CBs was higher in Chinese products than in global products (40.4% versus 25.2% for tri-CBs) (Breivik et al., 2002). Therefore, tri-CBs should be the dominant PCBs in the sandstorm depositions of Beijing.

Of the PCB homologues, the dominant PCBs detected in this study were indeed tri-CBs, accounting for more than 50.4% of the total PCBs in all sampling locations (Table 2). Higher-chlorinated homologues were detected at relatively low concentrations. The main PCB homologue in the average Chinese background and rural soils was also tri-CBs (55% and 38%, respectively) (Ren et al., 2007). The similarities of PCB patterns between sandstorm deposition and Chinese soil may indicate a common source origin.

3.3. PCA

We performed PCA to evaluate similarities or differences between the PCB homologue patterns of each sample; we normalized all data to percentage of Σ PCBs. PCs were determined by an eigenvalue of more than 1. By processing, two extracted PCs could explain 62.4% of the data variance. A loading plot and score plot rotation were obtained after varimax. Component 1 in Fig. 2 accounted for 36.5% of the total variance and was characterized by higher-chlorinated homologues, whereas component 2 accounted for 25.9% and was characterized by lower-chlorinated homologues. The score plot (Fig. 3) indicated that the compositions of PCBs in the sandstorm deposition samples were similar, possibly originating from the same source. The PCBs found in Beijing could therefore be classified into one main group that was relatively highly contaminated by tri-CBs. We also observed a clear clustering of samples, showing that tri-CBs are indeed the dominant contaminant in the sandstorm deposition of Beijing. All these samples as well as the #1 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 potentially associated with the #1 commercial PCB through long-range transmission.

3.4. PCB distribution

Fig. 4 shows the distribution of Σ PCBs in the sandstorm deposition samples, with an increasing trend from northwest to east Beijing. It is interesting to explore a correlation between Σ PCBs and the minimum particle sizes of the sandstorm deposition samples. We analyzed the particle size distributions of the sandstorm deposition samples by using a Laser Particle Size Analyser; the data are shown in Table 3. A statistically significant correlation existed between the Σ PCBs and minimum particle sizes ($r=0.81$; Fig. 5). This finding indicated that the Σ PCBs were associated with the particle size distribution and that they were removed through the scavenging of particulates. Another possible reason is the influence of the northwest wind during the study period. As a result, PCBs could be absorbed by smaller atmosphere particles

Fig. 2. Two-dimensional principal-component loading plot obtained from the data correlation matrix of 13 sandstorm deposition samples in Beijing.

Fig. 3. Two-dimensional principal-component score plot obtained from the data correlation matrix of 13 sandstorm deposition samples in Beijing.

Fig. 4. Distribution of Σ PCBs in urban areas of Beijing (data units in the figure are nanograms per gram).

Table 3
Particle size distributions of sandstorm deposition samples

Sample No.	<Particle size (μm) (particle number%)		
	<d1 (10%)	<d2 (50%)	<d3 (90%)
1	11.3	34.6	80.3
2	11.2	33.3	73.1
3	12.6	38.7	110.1
4	10.6	39.0	104.9
5	9.0	35.8	100.4
6	10.2	31.7	69.5
7	13.78	39.9	88.3
8	12.0	37.5	83.8
9	12.1	35.8	82.0
10	9.2	32.7	85.4
11	9.8	36.8	102.5
12	11.7	40.6	114.6
13	11.1	37.0	118.9

Fig. 5. Correlation between minimum particle sizes and total PCBs of Beijing sandstorm deposition samples.

that mixed with the passing deposition before being transported and deposited in eastern areas of Beijing, resulting in higher concentrations.

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