Levels and distributions of polychlorinated biphenyls in sewage sludge of urban wastewater treatment plants

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Abstract

Concentrations of polychlorinated biphenyls (PCBs) have been measured in sewage sludge samples from 8 urban wastewater treatment plants in Beijing, China. The PCB congeners were analyzed by isotope dilution high resolution gas chromatography/high resolution mass spectrometry method. The concentration of PCBs ranged from 65.6 to 157 ng/g dry weight (dw), with a mean value of 101 ng/g dw. The dioxin-like PCB WHO-TEQs (World Health Organization–Toxic Equivalents) of the sludge were lower than 1 pg/g dw. Consequently, all the concentrations of PCBs in sludge samples were below the upper limit for land application according to the Chinese legislation law for agriculture use. The PCB homologue profiles in sludge samples were dominated by tri-CBs and tetra-CBs. Similar distributions have been found in one of the Chinese PCB commercial products. The patterns of dioxin-like and indicator congeners observed in this study were quite similar in all samples. The predominant congener for dioxin-like and indicator PCBs were PCB-118 and PCB-28, respectively, while PCB-126 had the highest TEQ value.

Key words: polychlorinated biphenyls; wastewater treatment plant; sewage sludge; distribution

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Introduction

The utilization approach of sewage sludge as fertilizer to agricultural land is considered as an economical way to use the high amounts of sludge produced by the wastewater treatment plants (WWTPs) in China. Usually, sludge is often regarded as a potential manure resource, mainly because it contains high proportions of organic matter and abundant plant nutrients. However, it also consists of heavy metals and various persistent organic pollutants (POPs) (Cai *et al.*, 2007; Eljarrat *et al.*, 1997; Lazzari *et al.*, 2000) which might have a negative impact on soil organisms and fertility. Because of their lipophilic nature, many POPs such as polychlorinated biphenyls (PCBs), polychlorinated dibenzo-*p*-dioxins and dibenzofurans (PCDD/Fs) and polychlorinated naphthalenes (PCNs) preferentially partition onto sludge during wastewater treatment. These organic pollutants enter the wastewater stream by direct or indirect release from households and industry or by atmospheric deposition on surfaces and then runoff into the wastewater treatment system (Blanchard *et al.*, 2001; Katsoyiannis and Samara, 2004; Harrison *et al.*, 2006).

PCBs are a class of 209 chemical compounds that are ubiquitous in the environment. Among the PCB congeners, 12 have similar toxicity to that of PCDD/Fs are assigned with toxic equivalency factor (TEF) values by the World

Health Organization (WHO). Hence, these compounds are referred to dioxin-like PCBs. Additionally, 7 indicators have been selected to monitor PCBs to avoid the complexity involved in determining more congeners.

PCB commercial mixtures were widely used for different industrial applications such as dielectric fluids in capacitors and transformers, plasticizers in paints, adhesives, etc (Breivik *et al.*, 2002). Moreover, PCBs can be introduced into environment from waste incineration processes and occur as byproducts in a wide variety of chemical processes. In recent years, PCBs, especially dioxin-like and indicator congeners are of great environmental concern because of their high persistence, lipophilic properties, bioaccumulation, and toxicity to the ecosystem and human (Manodori *et al.*, 2006; Xing *et al.*, 2005). Because of these properties, PCBs have been banned since the end of 1970s in the USA and Europe and have been listed as serious threats to health and the environment by the Stockholm Convention (May 22, 2001). For these reasons, the concentrations, transport, fate and environmental risk of PCBs in various environmental media have been extensively studied (Pikkarainen, 2007; Manodori *et al.*, 2006; Zhao *et al.*, 2007; Howell *et al.*, 2008). Since sludge is widely recognized as an important matrix of persistent organic pollutants, including PCBs, this class of compounds has been well documented in other countries and regions (Souze Pereira and Kuch, 2005;

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Eljarrat *et al.*, 2003; Stevens *et al.*, 2003).

Many studies on the PCBs contamination in China were focused on the atmosphere (Lammel *et al.*, 2007), soils (Ren *et al.*, 2007), river sediments (He *et al.*, 2006; Liu *et al.*, 2007), biota (Zhao *et al.*, 2005), and human milk (Zhao *et al.*, 2007). However, little information on PCBs is available in the sludge of WWTPs in China. The main objectives of the present study are to investigate the levels and distribution of dioxin-like and indicator PCB congeners and total PCBs (from Mono-CBs to Deca-CB) in sewage sludge of WWTPs of Beijing and to identify their possible sources in sludge.

1 Materials and methods

1.1 Standards and reagents

All solvents were of pesticide residue grade and purchased from Tedia Co., USA. Silica gel (100–200 mesh) was obtained from Qingdao Haiyang Chemical Co., China. Permeable gel (SX-3 Bio-Beads, 200–400 mesh) was purchased from Bio-Rad Co., USA.

Calibration standard solutions, ^{13}C -labeled surrogate standards and injection standards complying with EPA method 1668A for PCBs analysis were purchased from Cambridge Isotope Laboratories, USA.

1.2 Sample collection and pretreatment

The sewage sludge samples were collected from WWTPs in Beijing in May, 2007, and their related details are shown in Table 1. Samples were packed in aluminum foil, then directly delivered to the laboratory and stored in a refrigerator. Sludge samples were freeze-dried for 48 h and mixed homogeneously before analysis.

1.3 Extraction and clean-up

The analysis of the PCB congeners was performed according to USEPA method 1668A. Approximately 2–3 g dry weight (dw) sludge samples was mixed homogeneously with appropriate anhydrous sodium sulphate in a glass thimble. The samples were spiked with a mixture of 1 ng ^{13}C -labeled PCB surrogate standards and Soxhlet-extracted for 24 h with toluene.

The concentrated extracts were subsequently purified

and fractionated by multi-layer silica gel, gel permeable column (GPC) and basic alumina chromatographic columns in turn (Guo *et al.*, 2008). A multi-layer silica column (from bottom to top: 1 g activated silica, 2 g AgNO_3 silica, 1 g activated silica, 3 g basic silica, 1 g activated silica, 8 g acidic silica, 1 g activated silica, and 2 cm anhydrous sodium sulphate) was used and eluted with 100 mL *n*-hexane. The second column was a gel permeable column packed with 30 g SX-3 Bio-Beads. Before loading the extracts, the column was pre-washed with 50 mL of 50% dichloromethane (DCM) in *n*-hexane. Samples were eluted with 120 mL of 50% DCM in *n*-hexane, the first 50 mL effluent was discarded and the following 70 mL effluent was collected. The extracts were further concentrated and passed through a column with 8 g basic alumina and 2 cm anhydrous sodium sulphate for further purification. Samples were then eluted with 100 mL of 5% DCM in *n*-hexane (PCBs and PCNs eluted in the same fraction). The final fractions were concentrated to 20 μL with gentle stream of nitrogen. Prior to injection, ^{13}C -labeled injection standards of PCBs were added for calculation of recovery.

1.4 Instrumental analysis

High resolution gas chromatography/high resolution mass spectrometry (HRGC-HRMS) measurements were carried out using an Agilent 6890 gas chromatography (GC) coupled with an Autospec Ultima mass spectrometer (MS) (Waters Micromass, UK) on a resolution of approximately 10000 with EI^+ mode (35 eV). HRMS was operated in selected ion monitoring (SIM) mode and the ion source temperature was 270°C.

A DB-5 (60 m $\times$ 0.25 mm $\times$ 0.25 μm film thickness) capillary column was used for the determination of PCB congeners. The column temperature initiated at 120°C (1 min), then increased to 150°C at 30°C/min, and 300°C (5 min) at 2.5°C/min. The injector and transfer line temperatures were 270 and 290°C, respectively. The carrier gas was helium at 1.2 mL/min. Injection volume was 1 μL in splitless mode.

1.5 Quality assurance and quality control

The procedures described above were controlled strictly by the analysis of procedural blank samples as well as the recoveries of ^{13}C -labeled compounds in each sample. Before extraction, the whole Soxhlet apparatus were pre-extracted for over 6 h with 250 mL toluene. Method blanks were monitored to evaluate the contamination. The results were free of PCB homologues except PCB-28 and PCB-52, which were 5% lower than the lowest sample concentrations. All sample concentrations in this study were not corrected with blank value.

The recovery in samples was obtained using the ^{13}C -labeled PCBs standards to ensure the repeatability of the results. Recoveries ranged from 62% to 114%. The limit of detection (LOD) for individual congener was defined by a signal to noise ratio of 3:1. The concentration of PCB congener in sample was destined for LOD value when it was lower than LOD value.

Table 1 Characteristics of the wastewater treatment plants (WWTPs)

Sample	Wastewater type	Process	Capacity (m^3/d)	Digestion treatment
S1	Domestic	OD	2.0×10^5	–
S2	Domestic	A^2/O	4×10^4	Aerobic
S3	Domestic	A^2/O	2.0×10^5	–
S4	Domestic	SBR	8×10^4	–
S5	Domestic	A^2/O	6.0×10^5	–
S6	Domestic	MBR	1.0×10^5	Anaerobic
S7	Industrial/Domestic (1:1)	A^2/O	5.0×10^5	Anaerobic
S8	Industrial/Domestic (1:1)	A^2/O	5.0×10^5	Anaerobic

OD: oxidation ditch; A^2/O : anaerobic-anoxic-oxic; SBR: sequencing batch reactor; MBR: membrane biological reactor.

2 Results and discussion

2.1 PCB concentrations in sludge samples

The analytical results of PCB concentrations for sewage sludge samples collected from 8 WWTPs in Beijing are presented in Tables 2 and 3. The total PCBs concentration (from Mono-CBs to Deca-CB) ranged from 65.6 to 157 ng/g dw with an average of 101 ng/g dw. Toxic equivalency (TEQ) concentrations of twelve dioxin-like PCBs are calculated using World Health Organization toxic equivalent factors (WHO-TEFs) (Van den Berg *et al.*, 1998). The WHO-TEQ values are not greater than 1 pg/g dw, which was far from the limit set at 100 pg TEQ /g dw for agricultural purposes (GB18918-2002). From the results of total PCBs, no correlation is observed between capacity or the treatment type of the WWTP and PCBs burden.

The high levels are detected at S7 (135.3 ng/g dw) and S8 (157.3 ng/g dw), both plants had a 50% input of industrial effluent, suggesting a possible industrial effluent discharge source. Similar influence has also been found on total polychlorinated naphthalenes (PCNs) of the same sludge samples (Guo *et al.*, 2008). According to the Chinese regulation (GB18918-2002), PCB levels should be less than 200 ng/g dw for agricultural use. Therefore,

the PCB values in sewage sludge in Beijing are lower than the maximum permissible concentration and are suitable for agricultural purposes.

The concentrations detected in this study are lower in sewage sludge from 14 wastewater treatment plants in UK (110–440 ng/g dw) (Stevens *et al.*, 2003), but are 1000-fold lower than in Brazilian sludge (57600–145000 ng/g dw) (Souze Pereira and Kuch, 2005), which might be explained the contamination of technical PCB mixtures.

As regards indicator PCBs, the total seven indicators PCB concentrations detected in sludge in Beijing (7.5–19.4 ng/g dw) are within the range reported in other similar studies which gave contents between 0.19 to 72.5 ng/g dw (Souze Pereira and Kuch, 2005; Eljarrat *et al.*, 2003). Blanchard *et al.* (2004) reported values for total indicator PCBs between 130 and 1070 ng/g dw, which is somewhat higher than the levels obtained in this study.

2.2 PCB homologue profiles and possible sources

Figure 1 presents PCB homologue profiles derived from sewage sludge samples. No significant changes have been found in all sludge samples. Tri-CBs (23.8%–46.9%) and Tetra-CBs (24.4%–44.3%) are the most dominant homologues of PCBs, followed by Di-CBs (6.7%–25.7%) and Penta-CBs (8.2%–13.6%). The contribution of other high-

Table 2 Concentrations of dioxin-like PCBs in sludges from WWTPs in Beijing, China (pg/g dw)

PCB congener	S1	S2	S3	S4	S5	S6	S7	S8
PCB-77	106	84.2	106	199	136	138	166	161
PCB-81	6.5	5.2	10.3	8.7	8.1	11.7	13.6	13.6
PCB-105	290	200	399	512	279	309	334	334
PCB-114	26.6	18.5	33.8	46.8	27.3	27.6	37.3	36.2
PCB-118	614	412	841	1046	585	657	690	699
PCB-123	52.8	41.8	76.5	97.1	52.3	54.6	72.1	73.0
PCB-126	3.6	3.3	3.6	7.1	4.5	3.6	7.6	7.2
PCB-156	60.4	44.7	105	106	63.3	63.2	80.4	79.2
PCB-157	14.2	11.1	24.4	26.2	15.4	14.3	19.1	19.6
PCB-167	20.4	15.8	36.3	39.6	23.4	22.1	32.0	31.1
PCB-169	0.8	0.7	0.8	1.5	1.1	0.8	2.5	2.4
PCB-189	3.4	3.3	5.2	6.1	3.9	2.9	13.4	12.6
Sum DL-PCBs	1199	841	1641	2096	1199	1305	1468	1469
WHO-TEQ	0.53	0.45	0.59	1.00	0.62	0.54	0.98	0.94

Table 3 Concentrations of indicator and homologue PCBs in sludges from WWTPs in Beijing, China (ng/g dw)

PCB congener	S1	S2	S3	S4	S5	S6	S7	S8
PCB-28	5.86	5.00	10.89	7.94	9.66	12.61	8.01	10.96
PCB-52	0.93	0.68	0.87	1.39	0.79	0.95	3.88	4.64
PCB-101	0.48	0.36	0.68	0.84	0.47	0.69	1.19	1.32
PCB-118	0.61	0.41	0.84	1.05	0.59	0.66	0.69	0.70
PCB-138	0.50	0.48	0.90	0.97	0.70	0.78	0.73	0.76
PCB-153	0.55	0.42	0.89	0.97	0.73	0.78	0.80	0.86
PCB-180	0.13	0.11	0.15	0.24	0.21	0.19	0.14	0.15
Sum indicator PCBs	9.07	7.46	15.22	13.39	13.16	16.67	15.44	19.39
Mono-CBs	0.06	0.08	0.05	0.29	0.23	0.05	0.06	1.93
Di-CBs	11.29	17.58	9.34	6.60	8.89	19.33	17.67	19.62
Tri-CBs	21.38	21.45	38.84	28.55	28.27	56.40	32.17	43.81
Tetra-CBs	30.17	20.93	20.21	43.76	18.23	29.27	34.09	38.91
Penta-CBs	7.77	5.59	9.03	13.42	5.41	10.89	13.63	14.73
Hexa-CBs	2.62	2.10	4.39	4.89	2.91	3.90	4.27	4.51
Hepta-CBs	0.36	0.30	0.41	0.67	0.42	0.62	1.54	1.56
Octa-CBs	0.23	0.19	0.26	0.40	0.28	0.30	5.38	5.09
Nona-CBs	0.04	0.03	0.04	0.06	0.06	0.03	4.20	4.35
Deca-CB	0.22	0.08	0.18	0.23	0.90	0.10	22.30	22.81
Total PCBs	74.13	68.32	82.75	98.86	65.60	120.9	135.3	157.3

Fig. 1 PCB homologue profiles of the sewage sludge samples.

chlorinated PCBs (from Octa-CBs to Nona-CBs) is less than 7% in all samples. The result reveals that PCB in sludge samples were predominated by low chlorinated PCBs. However, it must be noted that Deca-CB (PCB-209) has remarkably higher absolute concentrations and accounts for 15% in both S7 and S8 samples. As it is well known, because higher chlorinated homologues are less volatile and more bound to particles, they tend to resemble more accurately the estimated historical usage pattern and remain more closely to their sources regions (Meijer *et al.*, 2003). Consequently, the observed higher concentration of Deca-CB in S7 and S8 samples probably can be explained deposition into sewage sludge from the inputs of industrial effluent during the wastewater treatment processes. Because PCB-209 was not present in the most common PCB mixtures (Breivik *et al.*, 2002; Howell *et al.*, 2008), knowledge on distribution and sources of this congener is very limited. Howell *et al.* (2008) indicated that the unusually high fraction of PCB-209 was found from samples in Houston Ship Channel in Texas to unusual Aroclor mixtures used in the history or contemporary sources from local industry, while Ishikawa *et al.* (2007) confirmed PCB-209 was generated during the thermal process.

The observed homologue profiles relate reasonably well with PCB production. In China, about 10000 tons of PCBs were produced since 1965 to 1974, within 9000 tons as trichlorobiphenyl and 1000 tons as pentachlorobiphenyl (Xing *et al.*, 2005). Trichlorobiphenyl, known as PCB3,

was utilized in electric capacitor production and contained 42% chlorine, which had similar composition with Aroclor 1242. While pentachlorobiphenyl (PCB5), employed as additives in paint and contained 53% chlorine, was similar to Aroclor 1254 (Xing *et al.*, 2005; Jiang *et al.*, 1997). Figure 2 shows the homologue distribution in sewage sludge samples in the present study (Deca-CB was not taken into account) in comparison to the PCB technical mixtures. Compositions of PCB homologues in sewage sludge are in good agreement with those of Aroclor 1242. This indicates that the PCB3 products may be one of the major sources of PCBs in sewage sludge. The results are quite different with the results reported by Souza Pereira and Kuch (2005), in which both Brazilian samples show a homologue profile with the predominance of the penta-CB to hepta-CB groups that indicate a contamination from technical PCB formulations like Clophen A50 and Clophen A60 or equivalent.

2.3 PCB congener profiles

The profiles of dioxin-like and indicator congeners are quite similar in all samples regardless of difference of wastewater types, as shown in Fig. 3. This indicates that these PCB congeners influenced either by domestic or industrial input that have similar isomer compositions. Among the 12 dioxin-like PCB congeners, obviously, PCB-118, -105 and -77 were the predominant congeners (Table 2 and Fig. 3a), with the average contributions of 49%, 24%, and 10%, respectively. However, the PCB-TEQ values are exclusively dominated by PCB-126 (71% of the total PCB-TEQs) for it having a high TEF value. This observation agrees with the profile of dioxin-like PCB congeners found in sludges from Catalonia by Eljarrat *et al.* (2003). However, as regard to indicator PCB congeners, contribution of each congener has no significant difference in sludges from Catalonia (Eljarrat *et al.*, 2003), while the results of indicator PCB congeners (Fig. 3b, Table 3) in the present study show that PCB-28 was the most abundant congener, which accounts for approximately 65% of the total indicator PCBs.

Fig. 2 Distribution of PCB homologue groups (except Deca-CB) in sludges of WWTPs in composition with Aroclor commercial formulations (Aroclor 1242 and 1254)

Fig. 3 Profiles of dioxin-like (a) and indicator (b) PCB congeners in sludge of WWTPs

3 Conclusions

The present study reveals a moderate contamination of urban WWTPs in Beijing. Dioxin-like PCBs are detected in all the samples, but the TEQ levels are lower than 1 pg/g dw, which is far from the limit set at 100 pg/g TEQ dw for agricultural purposes. PCB homologue profiles are dominated by Tri-CBs, followed by Tetra-CBs and Di-CBs. The extremely high concentration of Deca-CB (PCB-209) detected in S7 and S8 samples might be due to the local industrial inputs. The observed homologue profiles relate reasonably well with one of the Chinese PCB commercial products. Dioxin-like and indicator PCB congener profiles in all samples are similar.

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