

Distribution of Polychlorinated Biphenyls (PCBs) and Toxic Equivalency of Dioxin-Like PCB Congeners in Rural Soils of Beijing, China

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ABSTRACT. The soil quality in rural area may associate with the health of local residents and food safety. This study aimed to investigate the spatial and vertical distribution of PCBs in rural soils of Beijing, and the source identification of PCBs and toxic equivalency calculation of dioxin-like PCB congeners were conducted. The mean of total PCB concentrations in Beijing rural soils was 11.01 ng g⁻¹. The congener profiles were predominated by lowly chlorinated congeners such as Di-CBs, Tri-CBs and Tetra-CBs. No significant difference of PCBs concentrations was observed between plain and mountain areas, and PCBs were slightly accumulated in the surface soils of Beijing rural area. Long-range transport of volatile PCB congeners was also an important source in addition to local sources of PCBs from electronic products. Toxic equivalency concentrations of dioxin-like PCB congeners met Canadian soil quality guidelines for agricultural soils, indicating a lower risk of PCBs to human health.

Keywords: polychlorinated biphenyls, rural soil, toxic equivalency, spatial distribution, Beijing

1. Introduction

Polychlorinated biphenyls (PCBs) are one of the most important persistent organic pollutants which were listed in Stockholm Convention in 2001. Industrial production of PCBs started in the UK in 1954, and more than 1.30 million tons of PCBs had been produced in the world till 1993 (Breivik et al., 2002a; Cachada et al., 2009). Due to their extraordinary chemical stability and heat resistance, PCBs are extensively employed as components in electrical and hydraulic equipment such as transformers, capacitors, hydraulic systems, etc. (USEPA, 1996). As a consequence, a large number of PCBs were released into the environment through a variety of ways (Breivik et al., 2002b; Liu et al., 2008). Many studies have reported the occurrence and levels of PCBs in soils, sediments, and air (Zhou et al., 2001; Garcia-Alonso and Perez-Pastor, 2003; Zhang et al., 2008; Batterman et al., 2009; Wu et al., 2011); and bioaccumulation and biomagnification were also observed in biota (Bureau et al., 2004; Millward et al., 2005). As we all know, PCBs can be persistently present in the environment, which increases their possibility of entering the human body (Longstaff, 2005). In addition, PCBs are of carcinogenicity and neurotoxicity (Calabrese and Sorenson, 1977; Hallgren and Darnerud, 2002). Therefore, PCBs have been banned in many countries.

However, so far, existing PCBs are still used as lubricant in many types of equipment, which may be a potential source of PCBs in environment.

Soils are an important reservoir for many contaminants including both inorganic and organic matters (Biasioli and Ajmone-Marsan, 2007; Aelion et al., 2009). Pollutant levels in soils should be given more attention due to their potential risk to public health through inhalation, diet and skin exposure. Fortunately, soils contaminated with PCBs have been concerned for many years (Meijer et al., 2003; Robson and Harrad, 2004; Heywood et al., 2006). The concentration, source, and congener pattern of PCBs in soils have been studied by many scientists all over the world. In China, PCB concentrations in soils of Dalian, Taiyuan, Linfen, Harbin, Hong Kong and so on have been reported in recent years (Zhang et al., 2007; Fu et al., 2008a; Wang et al., 2008; Fu et al., 2009; Ma et al., 2009). However, most of these studies focused on the concentration of PCBs in urban soils, only a few research reported the PCB levels in rural soils (Chen et al., 2008; Shen et al., 2009; Wang et al., 2009; Wu et al., 2010b). As well known, rural soils can provide abundant food and vegetables for local residents and urban area, and the soil quality may associate with food security. Therefore, an investigation of PCB distribution in rural soils in the vicinity of a large city with dense population such as Beijing is very important.

A preliminary study reported the levels and congener composition of PCBs in rural soils of Beijing, and correlations between PCBs concentrations and physicochemical properties of soils were also analyzed (Wu et al., 2010b). These results provi-

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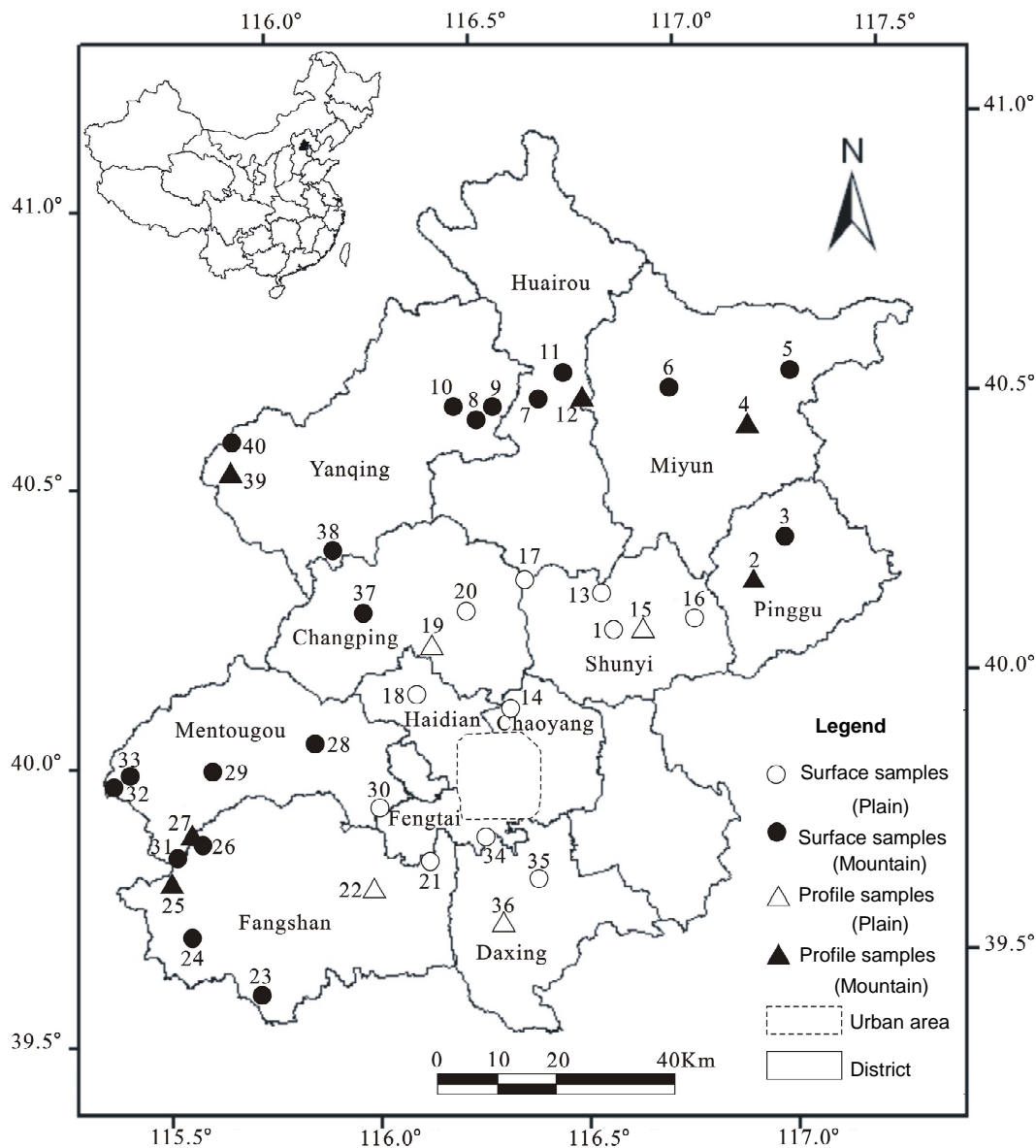


Figure 1. Sampling sites of rural soils in Beijing.

ded the basic information of PCBs in the rural soils of Beijing; however, further research is necessary to analyze the sources and assess the potential health risks of PCBs for developing a corresponding control strategy of PCBs contamination in rural soils of Beijing. The purpose of this study was to analyze the spatial and vertical distribution of PCBs in rural soils of Beijing, and explore their probable sources. In addition, toxic equivalency of dioxin-like PCB congener was calculated and compared with the soil quality guideline to provide more valuable information about the effect of PCBs level on human health.

2. Materials and Methods

2.1. Study Area

Beijing sits in the Northwest of North China Plain, with the latitude of 39.5 ~ 41.1 N and the longitude of 115.5 ~ 117.5 E

(Figure 1). It has a typical semi-humid continental monsoon climate, which is characterized by a hot, wet summer and a cold, dry winter. The annual mean temperature in Beijing is about 13.3 °C and the annual precipitation is about 481 mm in 2009. The area of Beijing is about 16,410 km² and the urban area is more than 1,000 km². The permanent population in Beijing is about 17.55 million, with a resident population density about 1,069 persons/km² in 2009. The soils of Beijing can be divided into seven soil groups, including mountain meadow soil, mountain brown earth, cinnamon soil, moisture soil, bog soil, paddy soil, and aeolian sandy soil.

2.2. Sample Collection and Pretreatment

Sample collection method has been described in previous paper (Wu et al., 2010a). The sampling criteria was based on

SEPA (2004), sampling sites were selected away from known or suspected contamination hot spots. Sampling sites (containing 30 topsoil sites and 10 soil profile sites) were distributed in 12 administrative regions, including 10 districts and 2 counties (Figure 1). A total of 60 soil samples were collected from the study area, including 30 samples from topsoil and 30 samples from soil profiles. For the topsoil samples, each sample was a mixture of five subsamples taken from the upper 20 cm in an area of 100×100 m. For the soil profiles, soil samples were collected from A (0 ~ 20 cm), B, and C layers, which represented leached horizon, illuvial horizon and parent material horizon, respectively. All soil samples were collected using a bamboo spade and stored in sealed kraft packages. The soil samples were dried in a shady place at room temperature; the impurities such as stones and tree leaves were removed from them. The samples were sieved to < 1 mm after drying and gently crushing aggregates without grinding sand-sized mineral fragments. Then a fraction of samples sieved to < 1 mm were ground with agate mortar to pass through a 0.15 mm sieve for the determination of PCBs.

2.3. PCBs Extraction and Cleanup

A total of 14 g soil sample was spiked with a recovery standard containing PCB 209, and then accelerated solvent extracted (ASE300, Dionex, America) with 30 ml of n-hexane/acetone (1:1, v/v) at 100 °C and 1500 psi. This process was repeated three times and the extracts were combined. Then the extracts were evaporated to 1 ml by rotary evaporator (RE-52, Shanghai Yarong Company, China), and cleaned using a chromatography column which consisted of the following: 2 g of silver nitrate silica, 1 g of activated silica gel, 3 g of basic silica gel, 1 g of activated silica gel, 4 g of acid silica gel, 1 g of activated silica gel, and 2 g of anhydrous sodium sulfate. PCB fractions were eluted with 100 ml of hexane. The collected eluant was concentrated and then reduced to 1 ml under a gentle N₂ stream for analysis.

2.4. GC-MS Analysis

A total of 18 PCB congeners were determined by GC-MS with electron impact ionization (Varian 4000, U.S.) (Table 1). Gas chromatographic separation was performed on a DB-5MS capillary column (30 m \times 0.25 mm \times 0.25 μ m film thickness) with a helium carrier gas. The injection was splitless mode. The column oven temperature was programmed at a rate of 10 °C min⁻¹ from an initial temperature of 80 °C (held for 2 min) to 160 °C and held for 1 min, 1.5 °C min⁻¹ to 230 °C and held for 15 min, 20 °C min⁻¹ to 280 °C and held for 15 min. Injector, transfer line, and ion source temperatures were 250, 280 and 250 °C, respectively. Selected ion monitoring (SIM) mode was used to acquire PCBs data.

2.5. Quality Assurance and Quality Control

All analytical procedures were monitored by strict quality assurance and quality control. Duplicates, blank samples and analysis of certified reference material were performed in each extraction batch of ten samples throughout all the experiments.

The target compounds were not detected in the blank samples. The detection limits for each PCB, estimated from the calibration curve as the concentration giving a blank signal plus three times standard deviations of the blank, were 0.01 ~ 0.06 ng g⁻¹ (dry weight). The matrix spike recoveries of 18 PCB congeners were 80 ~ 110%, with standard deviation being 14%. The recovery of PCB-209 surrogate in all samples was 80 ~ 105%. The calibration curves of PCBs were determined before each batch of samples, with the correlation coefficients (r) all greater than 0.99.

Table 1. Measured PCB Concentrations in Rural Soils of Beijing (ng g⁻¹ dry weight)

Congener	IUPAC number	Min	Max	Median	Mean	Std. Dev.
Di-CBs	8	<0.01	3.14	2.48	2.44	0.599
Tri-CBs	18	<0.01	1.95	<0.01	0.90	0.947
	28	<0.01	2.45	2.01	1.34	0.989
Tetra-CBs	44	<0.01	1.65	<0.01	0.41	0.709
	52	<0.01	1.63	<0.01	0.48	0.727
	66	<0.01	1.96	<0.01	0.62	0.894
	77	<0.01	2.60	<0.01	0.58	1.004
Penta-CBs	81	<0.01	2.07	<0.01	0.42	0.827
	101	<0.01	2.03	<0.01	0.36	0.771
Hexa-CBs	118/123	<0.02	1.69	<0.02	0.46	0.729
	128	<0.01	1.77	<0.01	0.51	0.775
	138	<0.01	1.60	<0.01	0.51	0.731
	153	<0.01	1.51	<0.01	0.53	0.714
Hepta-CBs	156/167	<0.02	1.54	<0.02	0.40	0.601
	180	<0.01	1.67	<0.01	0.58	0.744
	189	<0.01	1.56	<0.01	0.49	0.702
Σ 18PCBs		2.60	19.56	11.30	11.01	3.982

2.6. Statistical Analysis

Statistical analyses were conducted with Microsoft Excel (Microsoft Inc., USA) and SPSS 16.0 (SPSS Inc., USA). The distribution of PCB concentration was tested to determine if they approximated the normal probability function with the Kolmogorov-Smirnov method. Principal component analysis (PCA) was carried out for all the available samples and commercial mixtures of PCBs (Aroclor 1016, 1242, 1248, and 1254) to identify the possible sources of PCBs in the rural soils. The raw data were transformed to percentage concentrations for all soil samples prior to PCA, and the composition of PCB congener for each Aroclor mixture was obtained from USEPA (1996). The component matrix was rotated using a Varimax rotation to the axes, which maximized the variance of the components.

2.7. Calculation of Dioxin Toxic Equivalency for PCBs

Toxic equivalency (TEQ) concentration is a useful variable to evaluate the toxicity of dioxins and dioxin-like compounds, which has been applied to risk assessment of persistent organic pollutants such as PCBs (Schantz et al., 2003). In this paper, TEQ was adopted to estimate the human exposure and health risk of seven dioxin-like congeners (CB-77, 81, 118, 123, 156,

Table 2. Total Concentration of PCBs in Rural Soils of Beijing and Other Areas (ng g⁻¹)

Area	Category	PCBs			Reference
		Mean	Median	Range	
World	Background	5.41	---	0.026-96.9	Meijer et al., 2003
China	Rural	0.424	---	0.138-1.14	Ren et al., 2007
Dalian	Rural	1.337	1.337	---	Wang et al., 2008
Tibet	Rural	0.186	---	0.047-0.423	Wang et al., 2009
Hong Kong	Rural	0.11	0.10	0.07-0.16	Zhang et al., 2007
Great Britain	Rural	5.03	2.52	0.27-80.58	Heywood et al., 2006
Torino	Urban	---	14	1.8-172	Cachada et al., 2009
Madrid	Urban	32	---	9.0-66	Garcia-Alonso and Perez-Pastor, 2003
Moscow	Urban-rural transect	---	---	3.1-42	Wilcke et al., 2006
Beijing	Rural	11.01	11.30	2.60-19.56	This study

167, and 189) via dietary intake. The calculation was carried out as the following:

$$TEQ = \sum TEF_i \times c_i \quad (1)$$

where c_i was the concentration of the i^{th} PCB congener in rural soils of Beijing, and i was the IUPAC Number of PCB congener; TEF_i was toxic equivalency factor of the i^{th} PCB congener for human and mammals (Van den Berg et al., 2006). Then, the concentration of each dioxin-like PCB congener was converted to 2, 3, 7, 8-tetrachlorodibenzo-p-dioxin (TCDD) concentration.

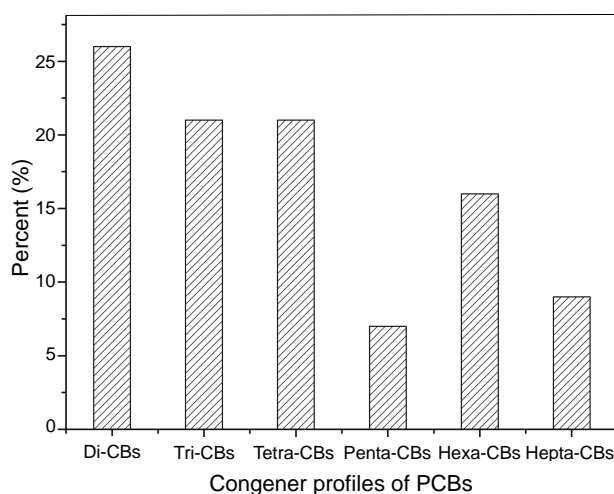
3. Results and Discussion

3.1. PCBs Levels in Soils

As a normal distribution was observed for the total concentrations of PCBs in rural soils of Beijing, parametric statistics was used to analyze all the data. Total concentration of PCBs in the rural soils ranged from 2.60 to 19.56 ng g⁻¹, with a mean of 11.01 ng g⁻¹ (Table 1). As to PCB congeners, the mean concentration of each homologue was less than 1.0 ng g⁻¹ except for PCB 8 and PCB 28. As shown in Table 2, the level of PCBs in rural soils of Beijing was higher than the mean concentration of PCBs in rural soils of China and their background value in the world. In comparison with other rural areas such as Dalian, Tibet, Hong Kong and Great Britain, the level of PCBs in rural soils of Beijing was also relatively higher. However, compared with urban area in Torino and Madrid and urban-rural transect in Moscow, the total PCB concentration was lower in rural soils of Beijing. As a conclusion, the total concentration of PCBs in rural soils of Beijing was higher than rural soils in other areas, but lower than those in urban area. This result may relate to the widespread application of PCBs in Beijing and the long-range transport and atmospheric deposition of volatile PCB congeners (Fu et al., 2008b).

3.2. Congener Profiles and Sources of PCBs

PCB profiles can reflect the fate of these contaminants; meanwhile it can be used to identify their sources combined with other techniques (Zhang et al., 2007). PCB homologues

**Figure 2.** Congener profiles of PCBs in rural soils of Beijing.

in rural soils of Beijing were predominated by Di-CBs (26%), which were followed by Tri-CBs (21%) and Tetra-CBs (21%) (Figure 2). Highly chlorinated congeners such as Hexa-CBs (16%) and Hepta-CBs (9.0%) were relatively low, and Penta-CBs (7.0%) were the least homologues. Overall, lighter chlorinated congeners including Di-CBs, Tri-CBs, and Tetra-CBs were dominant compositions of PCBs in Beijing rural soils, accounting for 68% of the total PCB concentration. This result was similar to the PCB homologues in rural soils of China, which was predominated by Tri-CBs (38%) followed by Di-CBs (25%) (Ren et al., 2007), suggesting that lowly chlorinated congeners may be an important source of PCBs in rural soils of Beijing.

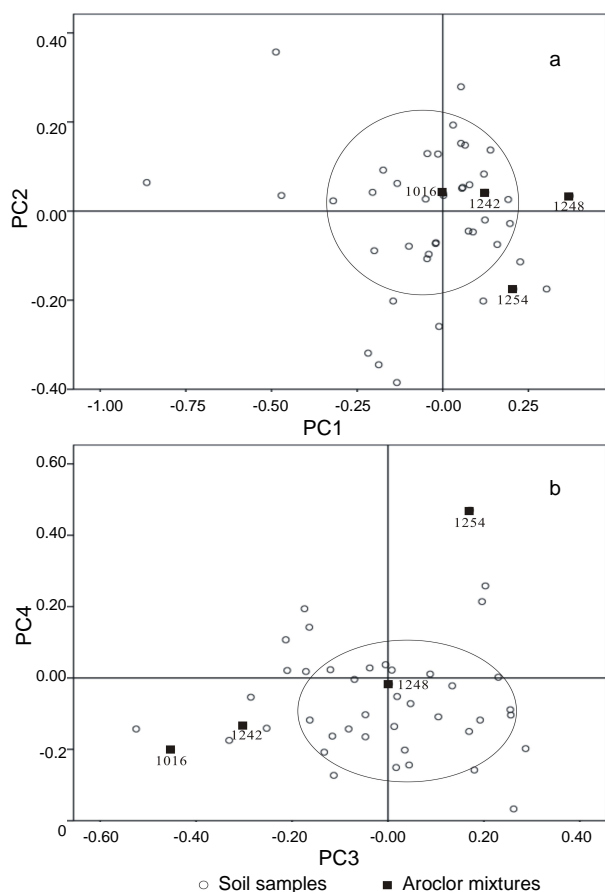
Since China began to produce PCBs in 1965, about 10,000 tons of PCBs including Tri-CBs (9,000 tons) and Penta-CBs (1,000 tons) had been manufactured until it was banned in 1974. Tri-CBs were primarily used in electrical products such as capacitor and transformer, while Penta-CBs were used as a paint additive (Xing et al., 2005). The common commercial mixtures of PCBs containing abundant Tri-CBs or Penta-CBs were Aroclor 1016, 1242, 1248, and 1254 (USEPA, 1996). Further sources identification of PCBs in Beijing rural soils was carried out by principal component analysis which combined all soil sam-

Table 3. Toxic Equivalency Concentrations (pg g^{-1}) of PCBs in Rural Soils of Beijing

Congener	TEFs*	Minimum	Maximum	Mean
77	0.0001	<0.001	0.260	0.058
81	0.0003	<0.003	0.621	0.125
118/123	0.00003	< 6×10^{-4}	0.051	0.014
156/167	0.00003	< 6×10^{-4}	0.046	0.012
189	0.00003	< 3×10^{-4}	0.047	0.015
$\Sigma 7\text{PCBs}^{**}$		<0.006	0.881	0.223

* Toxic equivalency factors obtained from WHO 2005 for human and mammals (Van den Berg et al., 2006).

** $\Sigma 7\text{PCBs}$ indicated the total concentration of seven dioxin-like PCB congeners including 77, 81, 118, 123, 156, 167 and 189.

**Figure 3.** Loading plot of principal component: (a) PC1-PC2; (b) PC3-PC4.

ples with these Aroclor mixtures. Four principal components (PCs with eigenvalue > 1) were extracted from the raw data, explaining 87.2% of the total variance (25.7, 24.5, 20.0 and 17.0%, respectively). PC1 referred to Di-CBs and Tetra-CBs, and PC2 referred to Hexa-CBs and Hepta-CBs. PC3 and PC4 were Tri-CBs and Penta-CBs, respectively. Loading plots were shown as to PC1-PC2 and PC3-PC4, respectively (Figure 3). As shown in Figure 3(a), Aroclor 1016, 1242 and most of the soil samples (28 soil samples, accounting for 70% of the total samples) could be classified into one group, indicating an iden-

tical source of PCBs. In addition, Figure 3(b) showed that Aroclor 1248 predominated by Tetra-CBs, Penta-CBs and Tri-CBs was another important source of PCBs in soil samples. Considering the history of industrial development in Beijing during the past several decades, the conclusion will be more clear and convincing. Since reform and opening, Beijing has undergone a rapid industrialization, especially in electronic industry. With the widespread use of electronic products, a large number of low molecular weight PCBs including Di-PCBs, Tri-PCBs and Tetra-PCBs with electronic waste has been released into the environment (Shen et al., 2009), which lead to higher concentration of lowly chlorinated PCBs in soils.

In addition, the highest relative content of Di-CBs (26%) in Beijing rural soils suggested that dechlorination mechanism may be a source of lowly chlorinated PCBs. The solubility and vapor pressure of highly chlorinated PCBs are lower, while their lipophilicity are higher than lowly chlorinated PCBs. As a consequence, more highly chlorinated PCBs are easier to accumulate in soils (Manz et al., 2001). Although the initial concentration of Tri-CBs and Penta-CBs released into Beijing rural soils might be relatively higher, Di-CBs became the predominant congener after a long time due to the dechlorination of Tri-CBs. In contrast, the content of Hexa-CBs (16%) and Hepta-CBs (9.0%) were higher than Penta-CBs (7.0%) due to their lower solubility and bioavailability.

Furthermore, high concentration of lowly chlorinated PCB congeners in Beijing rural soils may associate with the long-range transport of PCBs with low molecular weight. Many research indicated that volatile PCB congeners can spread to a long distance (Motelay-Massei et al., 2004; Primbs et al., 2008). Fu et al. (2008b) studied PCB residues in sandstorm depositions in Beijing, and found that Tri-CBs was the predominant homologues (> 50.4%). In summary, commercial mixtures such as Aroclor 1016, 1242, and 1248 as well as the long-range transport of volatile PCB congeners were the main sources of PCBs in rural soils of Beijing.

3.3. Spatial and Vertical Distribution of PCBs in Rural Soils of Beijing

Spatial distribution of PCB congener and PCBs in soils can reflect their possible sources (Motelay-Massei et al., 2004). The spatial distributions of PCBs in rural soils of Beijing were investigated in this study. According to the land terrain, rural area of Beijing was divided into two parts including plain and mountain. The mean concentrations of PCB congener in plain and mountain area ranged from 1.02 to 2.63 ng g^{-1} and from 0.68 to 2.49 ng g^{-1} , respectively. The total concentrations of PCBs in plain and mountain area were 11.09 and 10.96 ng g^{-1} , respectively. As shown in Figure 4, no significant difference was observed between plain and mountain area for the concentration of either individual PCB congener or total PCBs in rural soils of Beijing, implying an external input of PCBs was an important source in addition to local sources in Beijing rural area. This was in agreement with the results obtained in section 3.2.

Vertical distribution of PCBs in Beijing rural soils was also

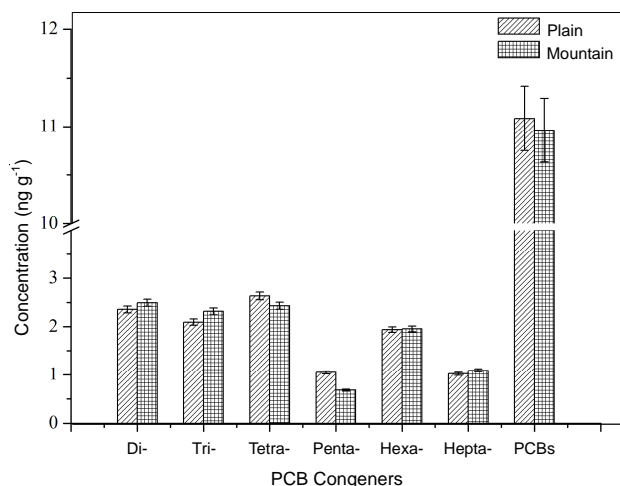


Figure 4. Spatial distribution of PCB congeners in rural soils of Beijing.

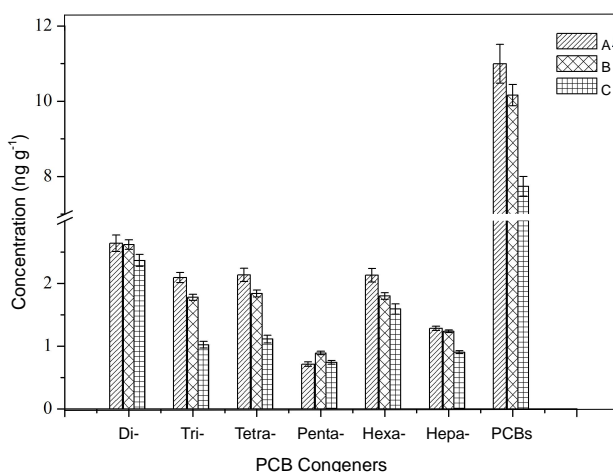


Figure 5. Vertical distribution of PCB congeners in rural soils of Beijing.

investigated. The results indicated that almost all PCB congeners and the total PCBs concentrations decreased from horizon A to C except penta-CBs, i.e. PCBs concentration in the topsoil was relatively higher (Figure 5). This was attributed to that PCBs come from anthropogenic sources and it transport from the topsoil to the subsoil (Chang and Li, 2008). In general, PCBs preferred to adsorb on organic matter (Ter Laak et al., 2009); in addition, higher content of organic matter was observed in the surface soils compared to subsoil in the rural area of Beijing (Wu et al., 2010a). Therefore, higher concentration of PCBs in surface soils was observed in rural soils of Beijing.

3.4. Toxic Equivalency of Dioxin-like PCB Congeners in Beijing Rural Soils

To evaluate the soil quality, Canadian soil quality guidelines were adopted. This standard was established to protect the environment and human health. The Canadian soil quality guideline of polychlorinated dibenzo-pdioxins/dibenzofurans (PCDD/Fs) for agricultural soils is 4 pg TEQ g⁻¹ (CCME, 2007).

The TEQ concentrations of dioxin-like PCB congeners in Beijing rural soils ranged from N.D. to 0.881 pg g⁻¹, with a mean of 0.223 pg g⁻¹ (Table 3). The total TEQ concentrations of all dioxin-like PCB congeners in all soil samples were lower than the Canadian soil quality guideline, indicating a safe soil environment for human health. However, due to their extreme toxicity to nervous system and biomagnification via food chain (Blankenship et al., 2005), PCBs levels in rural soils of Beijing should be paid more attention for their potential threat to human health.

4. Conclusions

The present work investigated the levels, congener profiles, spatial and vertical distribution, and sources of PCBs in rural soils of Beijing. The result indicated that the mean concentration of PCBs in Beijing rural soils was 11.01 ng g⁻¹. No significant difference of PCBs concentrations was observed between plain and mountain, and PCBs were slightly accumulated in surface soils of Beijing rural area. As to congener profiles, lowly chlorinated congeners such as Di-CBs, Tri-CBs, and Tetra-CBs were the dominant PCB homologues. Electronic products were important local sources of PCBs besides long-range transport of volatile PCB congeners. Although the levels of PCBs in Beijing rural soils were higher in comparison with other rural areas of China, PCBs concentrations in all soil samples met the available soil quality standards. Moreover, the total dioxin toxic equivalency concentration of all dioxin-like PCB congeners in each soil sample met the Canadian soil quality guidelines for the protection of environmental and human health.

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