



## Levels, distributions and profiles of polychlorinated biphenyls in surface soils of Dalian, China

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### ABSTRACT

Surface soil (0–5 cm) samples from 14 sampling sites including rural and urban areas of Dalian, a coastal city in Liaoning Province, China, were collected and analyzed for 84 polychlorinated biphenyls (PCBs). Total 57 PCB congeners were identified and mean concentration of total PCBs among all the sites was  $2.8 \mu\text{g kg}^{-1}$  dry weight (dw) with a range of 1.3 (rural site) to  $4.8 \mu\text{g kg}^{-1}$  dw (urban site). For the urban sites, total PCB concentrations in the soil samples collected in industrial area are highest, followed by those from business/residential sites and the garden sites, but the differences are not significant. Higher percentage of lighter weighted molecular PCBs were found in the rural site than urban sites in Dalian, possibly indicating the “urban fractionation effect”. Total seven dioxin-like PCBs (CB-77, 81, 105, 114, 118, 123, and 126) have been found in Dalian soils and the mean concentration (in  $\mu\text{g kg}^{-1}$  dw) among all the sites of CB-105 (0.17) is higher than any other dioxin-like PCBs, followed by that for CB-81 (0.14) and for 118 (0.06). No significant correlation relationship between the concentrations of dioxin-like PCBs and those of total PCBs was found. The toxic equivalency (TEQ) concentrations (in  $\text{ng kg}^{-1}$  dw) of these seven dioxin-like PCBs in 14 soil samples range from 0.006<sub>7</sub> to 4.885<sub>7</sub> (with a mean 1.372<sub>7</sub>) for humans and mammals, from 0.001<sub>7</sub> to 0.37<sub>7</sub> (with a mean 0.14<sub>7</sub>) for fish, and from 0.01<sub>7</sub> to 31.47<sub>7</sub> (with a mean 17.23<sub>7</sub>) for birds.

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### 1. Introduction

Polychlorinated biphenyls (PCBs) are highly persistent, toxic, bioaccumulative and widespread environmental contaminants, which are typical persistent organic pollutants (POPs) in the environment (Halsall et al., 1995). PCBs had been widely used for industrial purposes such as dielectric fluids in electrical transformers, capacitors, hydraulic and heat transfer fluids, and plasticizers. PCBs can be released from commercial PCB products, which are the potential primary source of PCBs in the environment (Herrick et al., 2007). Due to their toxic effects in both humans and wildlife (Cogliano, 1998; Giesy and Kannan, 1998), the levels and distributions of PCBs have aroused great concerns on environmental health.

Soils are natural sinks and environmental reservoirs for POPs in the terrestrial environment and thus understand of the fate of PCBs deposited to the surface soils is important. Being lipophilic compounds, PCBs adsorb to the organic carbon of the soil, and due to the high persistence and low mobility, PCBs can accumulate and

remain in the soil for very long periods of time. Atmospheric transport from major urban areas can lead to significant PCBs loading to surrounding terrestrial and aquatic ecosystems (Harner et al., 1995, 2004; Ockenden et al., 1998). Even today, many years after restrictions of PCBs manufacture and use were introduced in many countries, urban areas continue to act as major sources of atmospheric PCBs to surrounding regions (Halsall et al., 1995). It has been generally observed that PCB concentrations were higher in urban soils than in other soils (Wilcke et al., 2006; Jamshidi et al., 2007).

Therefore, the soil of urban areas is an important contamination source of PCBs. A number of studies have been conducted on the pollution of PCBs in environmental media such as atmosphere (Bi et al., 2002; Lammel et al., 2007), sediments (Zhou et al., 2001; Mai et al., 2005; Fu and Wu, 2006), and water (He et al., 2006) in China. However, few of the studies surveyed PCBs contamination in the soil, and when undertaken, the focus was on already heavily polluted areas (Xing et al., 2005). Recently, Ren et al. (2007) have for the first time investigated levels and distributions of PCBs in Chinese surface soil on a national scale. They found the average concentration of total PCBs among 52 sites (4 background, 39 rural, and 9 urban) across China in 2005 was  $515 \text{ ng kg}^{-1}$  dry weight (dw), approximately one-tenth of that in global background soil in 1998 (Meijer et al., 2003). The major PCB homologue group in

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Chinese background/rural soil is tri-PCBs followed by di-PCBs, while the major PCB homologue group in global background soil is hexa-PCBs followed by penta-PCBs (Meijer et al., 2003). These results show that PCB pollution in China have different character in comparison to other countries. Although PCBs levels of 9 urban sites were investigated in Ren et al. (2007) study, which carry urban signatures for the PCB congener profile, only one soil sample was collected in each city, not representative regarding the PCB concentration in surface soil in the city on a city-level scale. It is necessary to further investigate the contamination of PCBs and to identify the pattern of PCB profiles in Chinese urban areas.

The main objective of this study is to determine the distributions, concentrations and profiles of the PCBs in soils of China urban areas. Dalian (E120°58'–123°31', N38°43'–40°10') was selected as a typical coastal city, which situated in northeastern monsoonal area of China. The population in the urban area is more than 3.7 million, and total urban area of Dalian is about 2400 km<sup>2</sup>. The main industries include chemical engineering, shipbuilding, locomotive-building, tourism, and software development.

## 2. Materials and methods

### 2.1. Sample collection

In sampling place, a field about 100 × 100 square meters area was selected. Soil samples (0–5 cm) were collected at the four corners and the center of sampling fields and then the samples were spread onto a sheet of acetone rinsed aluminium foil and well mixed, from which, a certain amount (approximate 0.5 kg) of soil sample was taken. A total of 14 surface soil samples were collected at the area of the city of Dalian (see Fig. 1) in January, 2007, among which, four were industrial sites (9#, 11#, 12# and 13#), seven were business/residential sites (1#, 2#, 5#, 6#, 7#, 8#, and 10#), two were garden sites (3# and 4#), and one rural site (14#). All the samples sites are in urban areas except one (14#) in rural area, which is around 100 km north of Dalian urban areas. The samples were packed in a solvent-rinsed glass bottle with Teflon-lined cap, sent to the laboratory of the International Joint Research Center for Persistent Toxic Pollutants (IJRC-PTS), Dalian Maritime University, Dalian, China and stored at –20 °C until required for extraction.

### 2.2. Analytical procedure

Samples were treated, extracted, and analyzed according to the methods established at the National Laboratory for Environmental

Testing (NLET), Environment Canada. Three groups of PCB standards were purchased from the Accustandard, Inc. (New Haven, CT), Catalog numbers C-IADN-01, 02, and 03 containing a total 84 PCB congeners. The corresponding 84 PCB congeners can be found in previous study (Ren et al., 2007). There are 10 dioxin-like PCBs (CB-77, 81, 105, 114, 118, 123, 126, 167, 156, and 169) in these PCB congeners. Five grams of soil were isolated for percent moisture determination, and 20 g of each soil sample was accurately measured into a pre-cleaned extraction thimble and spiked with a recovery standard containing CB-65 and 155. The soil samples were then Soxhlet extracted for 24 h with 200 ml mix solvent (*n*-hexane/acetone, 1:1 v/v). The extract was cleaned up using a column filled with anhydrous sodium sulfate and silica gel. The column was made up of 10.0 g silica gel (activeness for 16 h at 135 °C) in lower part and 5.0 g anhydrous sodium sulfate (dryness for 8 h at 500 °C) in upper part. The column was washed with 30 ml hexane and 30 ml dichloromethane immediately prior to use. The extract was passed through the column, eluted with 50 ml mix of hexane and dichloromethane (1:1, v/v). The elution was rotary-evaporated to 2 ml and then reduced to 0.1 ml under a gentler nitrogen gas. Thirty-six nanograms of CB-30 and 204 were added finally as internal standard prior to analysis, respectively. All PCBs were identified and quantified with GC–MS (Finnigan PolarisQ). The mass spectrometer was operated in electron impact mode (70 eV). Helium gas was used as a carrier. The GC column was an HP-5MS fused silica column (60 m × 0.25 mm i.d., 0.25 μm film thickness). The column oven temperature was programmed at a rate of 10 °C min<sup>-1</sup> from an initial temperature of 80 °C (2-min hold) to a temperature of 160 °C (1-min hold), 1.5 °C min<sup>-1</sup> to 230 °C (15-min hold), 20 °C min<sup>-1</sup> to a temperature of 280 °C (10-min hold). Injector, transfer line and ion source temperatures were 250, 280 and 250 °C, respectively. PCBs were monitored in selected ion monitoring (SIM) mode at *m/z* = 222, 224 (di-PCBs), 256, 258 (tri-PCBs), 290, 292 (tetra-PCBs), 326, 328 (penta-PCBs), 360, 362 (hexa-PCBs), 394, 396 (hepta-PCBs), and 428, 430 (octa-PCBs).

### 2.3. Quality assurance/quality control

All samples were spiked with a labeled recovery standard (CB-65 and 155) prior to extraction. Sample recoveries averaged 89 ± 5% for CB-65 and 89 ± 10% for CB-155 in all samples. Spike samples were included at a rate of one for every seven soils extracted, and the average recoveries of all 84 PCBs are from 78% to 120%. The method detection limit (MDL) of 84 PCBs ranges from

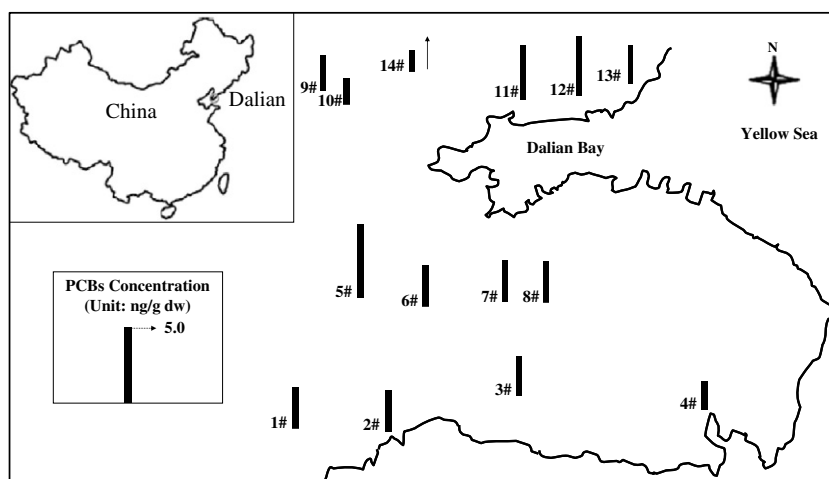


Fig. 1. The levels and the spatial distributions of  $\Sigma$ PCBs in surface soil of Dalian. Site 14# is a rural site, which is 100 km north of Dalian urban area.

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