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Size distribution and sorption of polychlorinated biphenyls during haze episodes

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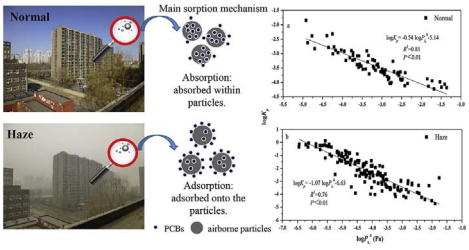
GRAPHICAL ABSTRACT



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ABSTRACT

There is a lack of studies on the size distribution of polychlorinated biphenyls (PCBs) during haze days, and their sorption mechanisms on aerosol particles remain unclear. In this study, PCBs in particle-sized aerosols from urban atmospheres of Beijing, China were investigated during haze and normal days. The concentrations, gas/particle partitioning, size distribution, and associated human daily intake of PCBs via inhalation were compared during haze days and normal days. Compared with normal days, higher particle mass-associated PCB levels were measured during haze days. The concentrations of ΣPCBs in particulate fractions were 11.9–134 pg/m^3 and 6.37–14.9 pg/m^3 during haze days and normal days, respectively. PCBs increased with decreasing particle size (> 10 μm, 10–2.5 μm, 2.5–1.0 μm, and ≤1.0 μm). During haze days, PCBs were overwhelmingly associated with a fine particle fraction of ≤1.0 μm (64.6%), while during normal days the contribution was 33.7%. Tetra-CBs were the largest contributors (51.8%–66.7%) both in the gas and particle fractions during normal days. The profiles in the gas fraction were conspicuously different than those in the PM fractions during haze days, with di-CBs predominating in the gas fraction and higher homologues (tetra-CBs, penta-CBs, and hexa-CBs) concurrently accounting for most of the PM fractions. The mean-normalized size distributions of particulate mass and PCBs exhibited unimodal patterns, and a similar trend was observed for PCBs during both days. They all tended to be in the PM fraction of 1.0–2.5 μm. Adsorption might be the predominating mechanism for the gas-particle partitioning of PCBs during haze days, whereas absorption might be dominative during normal days.

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1. Introduction

Atmospheric particulate matter (PM) has been a severe problem globally owing to its potential adverse health impact on humans. In the past few years, many epidemiological studies have proven that airborne PM is closely associated with increased mortality or morbidity (Hou et al., 2015), such as cardiovascular diseases, chronic respiratory illnesses, and lung cancer (Loomis et al., 2013; Pope III et al., 2002). The outdoor air pollution and PM from outdoor air pollution were recognized as a level 1 carcinogens to humans by the International Agency for Research on Cancer (IARC) Working Group (Loomis et al., 2013). The atmospheric fine particles, $PM_{1.0}$ and $PM_{2.5}$ (particles with aerodynamic diameter less than 1.0 μm and 2.5 μm, respectively), have aroused great attention. They can enter into the human body by respiration, based on characteristics of the tiny particle size. Studies have indicated that the fine particle fraction of < 0.5 μm may be the primary cause of the negative effects of particulate air pollution (Meng et al., 2013). Epidemiological evidence has also shown that mortality may be related to PM_{10}; the total daily mortality increases by about 1% for every 10 μg/m^3 increase in the concentration of PM_{10} (Lippmann, 1998).

Atmospheric fine particles may contain a variety of microorganisms (Cao et al., 2014), heavy metals (Hu et al., 2012; Pandey et al., 2013), and various organic pollutants (e.g., polychlorinated dibenzo-*p*-dioxins and dibenzofurans (PCDD/Fs), polybrominated dibenzo-*p*-dioxins and dibenzofurans (PBDD/Fs), polycyclic aromatic hydrocarbons (PAHs), and polychlorinated biphenyls (PCBs)) (Bi et al., 2005; Chrysikou and Samara, 2009; Okonski et al., 2014; Yang et al., 2013; Yue et al., 2015; Zhang et al., 2015; Zhu et al., 2016); therefore, ubiquitous fine particles associated with toxic pollutants are considered to be a great potential threat to humans. Hence, information on the particle size distribution of PM components have become more important for assessing the human risk of PM in the atmosphere.

Among the PM components, PCBs attracted great concern because some of these compounds are mutagenic, carcinogenic, bioaccumulative, persistent in the environment. The 12 dioxin-like PCB congeners are considered to be closely related to adverse human health, such as disruption of the endocrine system and carcinogenicity (Bhavsar et al., 2008). Though the production and use of PCBs were banned years ago, emissions of PCBs from a number of industrial thermal processes have been widely reported, including coking processes, waste incineration, and nonferrous metal production (Liu et al., 2013). PCBs have been found in the atmosphere around the world, including Algeria (Moussaoui et al., 2012), Venice (Gregoris et al., 2014), Japan, China, and Korea (Hogarh et al., 2012) in addition to polar regions (Baek et al., 2011; Cabrerizo et al., 2017). The gas–particle partitioning of PCBs in the atmosphere has been widely reported (Castro-Jiménez et al., 2009; Kim et al., 2011; Wang et al., 2017). Nonetheless, in most cases, it still remains unclear whether adsorption or absorption is the dominating mechanism, owing to varying ambient conditions. To better understand the size distribution of PCBs in the atmosphere, several research groups have analyzed PCBs in airborne particles (Chen et al., 1996; Chrysikou et al., 2009). However, the information regarding PCB fractionation in different size particles during haze days is still insufficient. Thus, knowledge of the sorption mechanisms and size distribution during haze days is essential, which will help to understand their environmental behavior in the atmosphere.

Beijing is an international metropolis with a high dense population, heavy traffic, and rapid economic growth, which has resulted in frequent and severe haze events in recent years (Ouyang, 2013; Tian et al., 2014). It was reported that a significant increase of respiratory-related outpatient cases have occurred because of massive haze events (Cao et al., 2014). PM has become a major problem in Beijing at present, which may pose a formidable public health threat. Much attention has been paid to PCBs in the atmosphere. The concentrations of PCBs in the atmosphere in Beijing have been reported (Xu et al., 2005; Li et al., 2009), but the size distributions of particulate-bound PCBs during the frequent and severe haze events that occur in Beijing have not been clarified. PCB concentrations may be elevated during haze days, and the potential health risks posed to humans exposed to PCBs will be more severe during haze days than during normal days. It is therefore important that the particle size distributions of PCBs in Beijing in extreme air conditions are investigated.

In previous papers, we reported general distributions of PCBs in the atmosphere (Zhu et al., 2017). Here, we have focused on better understanding the variation of atmospheric PCB concentrations, gas-particle partitioning, and homologue profiles in the atmosphere in Beijing, China during haze days and normal days. The size distributions of PM-associated PCBs were investigated. Furthermore, to determine if the haze days will show measurable differences in risk assessment via inhalation, the human inhalation exposure of PCBs in different size airborne particles was also evaluated.

2. Materials and methods

2.1. Air sampling

Sample acquisition was carried out from June to November 2014 with the high volume cascade impactor (KS-303 p.m.$_{10/2.5/1.0}$, Kálmán System, Hungary). The sampling location was at the Beijing Urban Ecosystem Research Station (116°12′28″ E, 40°00′44″ N), which is regularly used for air monitoring studies. High volume air samplers were used to collect the air samples. Each sample was collected for 3–6 d at a constant flow rate of 24 m^3/h. During the sampling period, twelve group air samples were collected and totally 60 samples were analyzed as every collected sample included five samples (a gas phase fraction and 4 p.m. fractions: > 10 μm, 10–2.5 μm, 2.5–1.0 μm, and ≤1.0 μm). The quartz fiber filters (QFFs) were used to collect PM fractions. Each QFF was weighed before and after a sample was collected using an analytical balance with a precision of 0.0001 g. The QFF was equilibrated in a chamber at a constant temperature (20 ± 5 °C) and humidity (30% ± 2%) before it was weighed.

Air samples were divided into haze samples and normal samples according to the $PM_{1.0}$ concentrations and visibility in this study. Haze samples were collected when $PM_{1.0}$ concentrations were > 100 μg/m^3 and visibility was < 10 km. (Lin et al., 2014; Zhang et al., 2016). Detailed sampling information (date, air volumes, and average temperature) is listed in Table 1.

2.2. Sample extraction and analysis

Full details of the analytical procedures have been reported in previous studies (Hu et al., 2014; Zhu et al., 2017). Briefly, PCBs were analyzed by the isotope dilution high-resolution gas

Table 1
Sampling and meteorological condition.

Sample name	Sample number	Sampling date (d/m/yyyy)	Volume sampled (m^3)	Temperature (°C)
Normal	N1	21–26.6.2014	3045	25.0
Normal	N2	26.6–2.7.2014	3851	27.6
Normal	N3	7–10.8.2014	1867	26.4
Normal	N4	6–9.9.2014	1629	23.9
Haze	H1	9–12.10.2014	3456	16.1
Haze	H2	18–20.10.2014	2310	16.0
Haze	H3	22–24.10.2014	2308	13.8
Haze	H4	13–15.11.2014	2295	5.2
Haze	H5	19–21.11.2014	2312	5.5
Haze	H6	25–27.11.2014	2311	4.2
Haze	H7	23–30.12.2014	3433	1.1
Haze	H8	4–9.1.2015	3445	1.0