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Organohalogen contamination in passerine birds from three metropolises in China: Geographical variation and its implication for anthropogenic effects on urban environments

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ABSTRACT

Contamination of organohalogen pollutants (OHPs), including dichlorodiphenyl trichloroethane and its metabolites (DDTs), polychlorinated biphenyls (PCBs), polybrominated diphenyl ethers (PBDEs), decabromodiphenylethane (DBDPE), hexabromocyclododecanes (HBCDs), and dechlorane plus (DP) in three metropolises of China, Beijing, Wuhan, and Guangzhou, and a reference rural site were determined using terrestrial residential passerine species as bioindicator. DDTs dominated in Wuhan whereas flame retardants dominated in Guangzhou and Beijing. No geographical variation was found for PCB levels but it exhibited different homologue profiles among different sites which could be attributed to different dietary sources of birds. Industry characteristics of the sampling location contributed to the geographical differences in the occurrence and contamination profile of OHPs. The transformation of traditional agriculture characterized contamination profiles to industry characterized profiles in Beijing and Guangzhou implicates significantly environmental concern on the flame retardants contamination in non-hot-spot regions of China.

1. Introduction

The occurrence of organohalogen pollutants (OHPs), such as polychlorinated biphenyls (PCBs), dichlorodiphenyl trichloroethane and its metabolites (DDTs), polybrominated dipheny lethers (PBDEs), decabromodiphenylethane (DBDPE), hexabromocyclododecane (HBCDs), and dechlorane plus (DP), has been of great concern because of their persistent, bioaccumulative nature, and their adverse effects on both humans and wildlife. Numerous studies have demonstrated ubiquitous OHP contamination in biota and abiota in China (Law et al., 2008; Ma et al., 2012; Wu et al., 2012). By contrast with the modest growth in the European and North American market, the demand and consumption of flame retardants (FRs), including PBDEs, DBDPE, HBCDs, and DP, is still experiencing a rapid rise with the annual growth of nearly 10% in China (The Freedonia Group, 2011). Legacy pollutants, involving PCBs and DDTs, have been continuously releasing into the Chinese environment due to historical usage and emission (Zhang et al., 2002).

Passerine birds have been successfully used in OHP biomonitoring in terrestrial ecosystems (Eens et al., 2013; Sun et al., 2012a; Van den Steen et al., 2009). Residential passerine species are especially suitable to reflect local contamination because of their small home ranges, territories, and foraging areas (Dauwe et al., 2006; Van den Steen et al., 2008). Moreover, terrestrial passerine birds with large populations are widespread and easily collected, which enables large geographical-scale monitoring and international intercomparison on OHPs pollution (Van den Steen et al., 2009).

The Eurasian tree sparrow (*Passer montanus*, ETS), which is characterized as human commensals (Li et al., 2008), is generally exposed to OHPs via the similar routes (e.g. inhalation and food) as humans. It is also a common bird species with a broad distribution in varieties of environments along urban gradient in China (Zhang and Zheng, 2010). Accordingly, they are particularly suitable model species to study the effects of urbanization closely linked to human

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living environments (Zhang and Zheng, 2010). As for the common magpie (*Pica pica*, CM), it is also a common songbird in most of the northern hemisphere (Jaspers et al., 2008). Throughout its distribution range, the magpie is associated with human activities (Jerzak, 2001). Previous studies have demonstrated synurbanization of this species, owing to its abilities to exploit anthropogenic food resources and to nest near human settlement (Jerzak, 2001).

In the present study, various classes of OHPs, involving DDTs, PCBs, PBDEs, DBDPE, HBCDs, and DP, were determined in two terrestrial passerine bird species, ETS and CM. Beijing, Wuhan and Guangzhou were selected as targeted study areas based on their representativeness in the corresponding geographic position of China from North to South (Fig. S1). The objective of the present study was to investigate the spatial distribution in the occurrence of different OHPs and to obtain the comprehensive contamination profiles in the Chinese urban environment. This study would be significative to extend the database about background levels of OHPs in non-hot-spot regions, i.e. e-waste recycling areas, of China, to explore the local usage and emission of OHPs, as well as to provide a baseline for further long-term biomonitoring in larger geographical scale throughout China.

2. Materials and methods

2.1. Sampling

During the non-breeding season, a total of 76 birds, including 67 ETS and 9 CM, were collected between August 2009 and May 2011 from three regional metropolises and a reference site in China (Fig. S1). Of the three cities, Beijing (BJ), located in the North China Plain, is the capital city of China. 40 ETS were collected from 8 sites located in the downtown areas containing ~7500 people per km^2 of Beijing City. The sampling sites included residential communities, campus, park, and industrial area. Wuhan (WH), situated at the midstream of the Yangtze River, is the largest metropolis in the Central China and has been one of the most important bases of heavy industry and cotton origin over decades. 9 CM were collected from 1 site in the suburb area with the human population density of ~500 people per km^2. Guangzhou (GZ), as the principal capital of Guangdong Province, is a highly industrialized and urbanized city in the Pearl River Delta of South China. 19 ETS were collected from 2 sites in the downtown areas with a mean human population density of ~14 000 people per km^2. One site located in a campus and one site located in a residential community. Shaoguan (SG), located in the northern Guangdong Province, was characterized by agricultural activities with less population density (~100 people per km^2 in the sampling site) and therefore was chosen as a reference site in the present study. 8 ETS were sampled from a rural site located in Shaoguan (Table 1). The details of avifaunal sampling method were given in Zhang et al. (2011). The necessary permit for the sample collection was obtained from the local Forestry Bureau. Birds were transported immediately to the laboratory and euthanized with N_2. Pectoral muscle was excised from each specimen and stored at −20 °C prior to chemical analysis.

2.2. Sample preparation and analysis

Procedures for sample pretreatment have been given in details in our previous study (Yu et al., 2011) with modest modifications. Approximately 1 g muscle tissue was homogenized after lyophilization and Soxhlet extracted with acetone/hexane (1:1, v/v). Surrogates (BDE-77, -181, ^{13}C-BDE-209 for PBDEs, DBDPE and DP, CB-30, -65, and -204 for PCBs and DDTs, ^{13}C-labled α-, β-, and γ-HBCD for HBCD diastereoisomers, respectively) were spiked prior to extraction. An aliquot of the extract was used for gravimetrical determination of lipid content. The remainder was further cleaned through gel permeation chromatography (GPC), followed by cleanup on a 2 g silica gel solid phase extraction column (Isolute®, Biotage AB, Sweden). The fraction containing all target compounds was obtained by 6.5 mL hexane/dichloromethane (60:40, v/v) and another 7 mL dichloromethane. The purified extracts were spiked with internal standards (BDE-118 and -128 for PBDEs, DBDPE and DP, CB-24, -82, and -198 for PCBs and DDTs, d_{18}-labled α-, β-, and γ-HBCD for HBCDs, respectively) before instrumental analysis. PBDEs, DP and DBDPE were analyzed by gas chromatography-electron capture negative ionization mass spectrometry (GC/ECNI-MS) in selected ion monitoring (SIM) mode. For tri- to hepta-BDE congeners and DP isomers, a 30 m × 0.25 mm i.d. × 0.25 μm DB-XLB capillary column (J&W Scientific, CA) was used. For the determination of octa- to deca-BDEs and DBDPE, a 15 m × 0.25 mm i.d. × 0.10 μm DB-5HT capillary column (J&W Scientific, CA) was used. PCB congeners, as well as dichlorodiphenyl trichloroethane and its metabolites (DDTs), were separated on a DB-5MS column (60 m × 0.25 mm i.d. × 0.25 mm, J&W Scientific, CA) installed on an Agilent 6890 GC-5975 MS system equipped with electron impact (EI) ion source. An Agilent 1200 series liquid chromatography (LC) system coupled to an Agilent 6410 electrospray triple quadrupole mass spectrometer operated in electrospray ionization (ESI) negative ion mode was used for HBCDs determination. HBCD diastereoisomers were separated through an XDB-C_{18} column (4.6 mm i.d. × 50 mm, 1.8 μm, Agilent, CA). More details of instrumental analysis were given in the Supplementary Data.

Stable nitrogen and carbon isotope analysis (δ^{15}N and δ^{13}C) were conducted on a Flash EA 112 series elemental analyzer coupled with a Finnigan MAT ConFlo III isotope ratio mass spectrometer using freeze-dried subsamples. Specific analytical approach and the assessment of isotopic ratios were previously given elsewhere (Yu et al., 2011).

2.3. QA/QC and data analysis

Procedural blanks were consistently analyzed and the mean values were used for subtraction. HBCD diastereomers were non-detectable in blanks and only few PBDE congeners, such as BDE-47, -206, -207, -208, and -209, and DP isomers (<10% of that in the samples) were found in blanks. The mean (±standard deviation) recoveries in matrices spiking triplicates ranged from 82.1 ± 1.3% to 104.3 ± 8.0% for individual PBDE congeners, 67.7 ± 16.7% to 94.1 ± 2.4% for seven PCB indicators (CB-28, -52, -101, -118, -138, -153, and -180), and 114.8 ± 7.3%, 90.0 ± 11.7% and 62.3 ± 5.6% for α-, β- and γ-HBCD, respectively. The surrogate standard recoveries

Table 1
Median concentrations and ranges (in parentheses) of the investigated OHPs in the studied samples (ng/g lw).

	Reference site	Urban sites		
	Shaoguan (SG)	Guangzhou (GZ)	Beijing (BJ)	Wuhan (WH)
Species	ETS[a] (n = 8)	ETS (n = 19)	ETS (n = 40)	Common magpie (n = 9)
Lipid (%)[b]	12.0 ± 2.3	11.5 ± 4.4	10.4 ± 4.1	8.1 ± 1.9
δ^{13}C (‰)[b]	−24.7 ± 1.1	−20.2 ± 4.7	−18.7 ± 3.3	−23.8 ± 0.4
δ^{15}N (‰)[b]	8.7 ± 0.4	6.0 ± 1.4	6.8 ± 1.3	5.5 ± 1.2
ΣOHPs[c]	210 (150–3900)	430 (180–5900)	860 (250–13000)	1500 (690–19000)
ΣDDTs[d]	92 (38–3800)	150 (23–880)	340 (89–11000)	1400 (610–15000)
ΣPCBs[e]	44 (24–69)	88 (26–540)	59 (23–720)	87 (25–500)
ΣPBDEs[f]	28 (16–51)	190 (51–3700)	250 (100–2600)	32 (12–2900)
DBDPE	nd[i]	31 (2.8–390)	8.5 (nd–330)	3.6 (0.18–820)
ΣHBCDs[g]	nd	3.4 (0.68–21)	51 (6.5–1100)	2.8 (2.7–14)
ΣDP[h]	24 (8.9–120)	13 (nd–350)	4.9 (nd–31)	4.4 (1.7–18)

[a] ETS: Eurasian tree sparrow.
[b] Mean ± SD.
[c] Sum of ΣDDTs, ΣPCBs, ΣPBDEs, DBDPE, ΣHBCDs, and ΣDP.
[d] Sum of *p,p′*-DDE, *p,p′*-DDD, and *p,p′*-DDT.
[e] Sum of PCB-28/31, -52, -60, -66/95, -74, -99, -105, -118, -128, -130, -137, -138, -146, -149/107, -153/132, -156, -158, -163, -167, -170/190, -178, -180/193, -183, -187, -191, -194, -203, -206, -207, -208 and -209.
[f] Sum of BDE-28, -47, -99, -100, -153, -154, -183, -196, -197, -201, -202, -203, -206, -207, -208, and -209.
[g] Sum of α-, β- and γ-HBCD.
[h] Sum of *syn*-DP and *anti*-DP.
[i] Not detectable or below the MDLs.

from authentic samples were (mean ± standard error) ranged 82.9 ± 1.1%, 71.7 ± 1.6%, and 82.3 ± 3.4% for BDE-77, -181 and ^{13}C-BDE-209, 76.4 ± 1.4%, 77.1 ± 1.6%, and 77.5 ± 2.5% for CB-30, -65 and -204, 73.8 ± 2.9%, 72.8 ± 2.4%, and 60.8 ± 2.3% for ^{13}C-labled α-, β-, and γ-HBCD, respectively. The method detection limit (MDL) was defined as a mean value plus three times the standard deviation in procedural blanks (n = 10). For analytes which can not be detected in blanks, a signal-to-noise ratio of ten was set as corresponding MDL. MDLs were at the range of 0.10–9.8 ng/g lipid weight (lw) for tri- to nona-BDEs, PCBs, DDTs, HBCDs and DP, 29 ng/g lw for BDE-209, and 3.3 ng/g lw for DBDPE, respectively (see details in Supplementary Data, Table S1).

All concentrations were expressed on a lipid weight (lw) basis. ΣPBDEs, ΣPCBs, ΣDP, ΣHBCDs, and ΣDDTs are defined as the sum of 16 BDE congeners (BDE-28, -47, -100, -99, -154, -153, -183, -201, -202, -197, -203, -196, -208, -207, -206, and -209), 37 PCB congeners (Table 1), *anti*- and *syn*-DP, three HBCD diastereomers (α-, β- and γ-), and *p,p′*-DDE, *p,p′*-DDD and *p,p′*-DDT, respectively. Non-normally distributed data (determined by the Shapiro–Wilk test) were logarithmically transformed to normal distribution, before being subjected to one-way analysis of variance (ANOVA) accompanied by Tukey HSD tests on SPSS 17.0. The level of significance through the present study was set at $\alpha = 0.05$.

3. Results and discussion

3.1. Stable isotopic analysis in the reference versus the urban passerine birds

Stable carbon and nitrogen isotopes provide independent measurement of carbon source and trophic level in wildlife (Jardine et al., 2006). Samples from BJ and GZ exhibited wide range of δ^{13}C and δ^{15}N values compared to those from SG and WH (Fig. 1), indicating the high heterogeneity of diet source for birds in BJ and GZ. Generally, δ^{13}C values in samples from BJ and GZ were significantly higher than those in samples from SG and WH ($p < 0.005$ for δ^{13}C, $p < 0.001$ for δ^{15}N, Table 1, Fig. 1), which was in line with the peregrine study from California (Newsome et al., 2010a), where samples collected from urban areas have higher δ^{13}C than those from non-urban areas. Sparrows are predominantly seed and grain eating bird (Zhang and Zheng, 2010). The main components of the diet of sparrows therefore rely on the vegetation structure in their habitat. Regarding magpies, which are omnivores and preferred to forage on garbage, their diet composition is strongly linked to the availability of anthropogenic food (Jerzak, 2001). Due to the urbanization and industrialization, the cover rate of forest which characterized by C_3 plant decreased (Statistical Yearbook 2011 for Beijing, Wuhan and Guangzhou, respectively) and was replaced by secondary herbage based on C_4 photosynthetic pathway. This would enhance the δ^{13}C value (Bird et al., 1996). Newsome et al. also found heavier δ^{13}C in kit foxes from urban sites compared to those from nonurban areas (Newsome et al., 2010b). Six samples which were collected from a university campus (GZ2) planted with indigenous shrubs in GZ showed lower δ^{13}C values (Fig. 1), indicating that these samples have different diet source from the samples collected in the other urban sites of GZ. The reference site (SG) located in northern Guangdong was characterized by abundant forest with a forest coverage rate of 75%. The similar δ^{13}C values in birds collected from SG and the university campus site (GZ2) suggested that plants play an important role in the diet of passerine birds.

The δ^{15}N values of samples from SG were higher than those from other cities (Fig. 1), which may partly be explained by the different isotopic baselines as well as the variation in isotopic fraction among different species and ecosystems (Post, 2002).

3.2. Geographical variation of contaminant levels

Levels of ΣDDTs, ΣPCBs, ΣPBDEs, DBDPE, ΣHBCDs, and ΣDP in the birds from the reference site (SG) were generally lower than that from the urban sites (Table 1), which was in line with the expectation of less contamination of OHPs in the rural areas. *p,p′*-DDE was the most prevalent compound, constituting more than 94% of the sum DDTs, which was in accordance with the dominance of *p,p′*-DDE in most passerine studies (Eens et al., 2013; Van den Steen et al., 2009; Van den Steen et al., 2010). Sum DDT concentrations differed significantly among the sampling locations ($F_{3,72} = 17.157$, $p < 0.0001$). WH burdened the highest DDT levels among the four sampling sites (Fig. 2), which could be related to the fact that Hubei Province where WH located is a major agricultural province characterized by abundant cotton planting in the past decades. DDTs had been heavily used in this province. Additionally, the sampling site in Wuhan is situated in the suburban areas whereas the sampling sites in GZ and BJ are located in the downtown areas. This may enlarge the contamination gap of DDT between WH and other two cities. No obvious geographic variation can be found for PCBs ($F_{3,72} = 2.382$, $p = 0.077$, Fig. 2), indicating a relative homogeneity for PCB level in China.

Significant differences for ΣPBDE concentrations were observed among the sampling locations ($F_{3,72} = 17.239$, $p < 0.0001$, Fig. 2). Levels in the reference site were significantly lower than those of GZ and BJ, which was generally linked to urbanization and industrialization (Jaward et al., 2004; Van den Steen et al., 2009). The per capita gross domestic product (GDP) in the reference site (~20 000 RMB in 2010), according to the statistics, is much lower than that of GZ and BJ (~80 000 and 70 000 RMB in 2010, respectively). The gross output value of the primary industry (i.e. agriculture) accounted for approximately 20% in GDP in the reference site, whereas the proportion of the primary industry in GDP was less

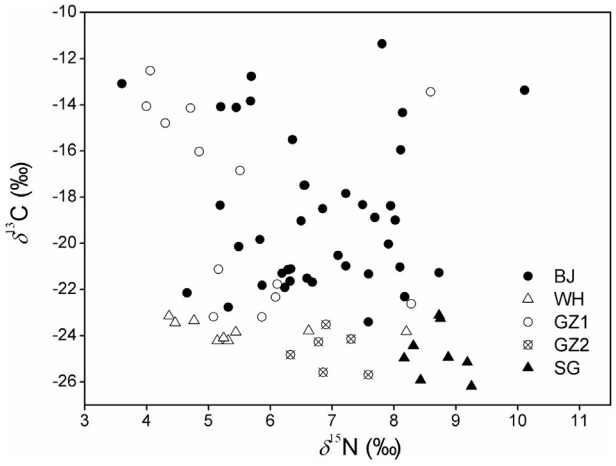

Fig. 1. Stable isotope ratios of carbon and nitrogen in muscle of passerine birds from different sampling sites. GZ2 was located in a university with high cover rate of C_3 plant which showed similar stable carbon isotopic composition to that in SG.

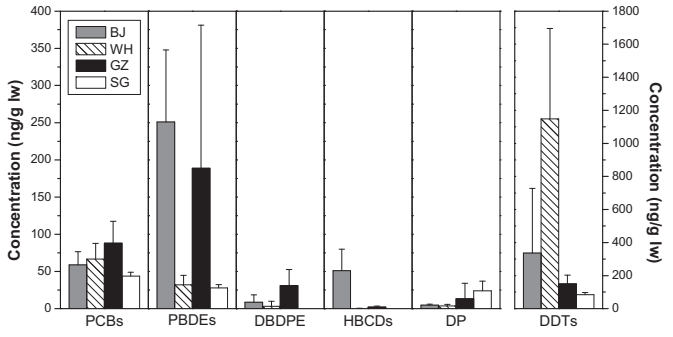

Fig. 2. OHP levels in muscle of passerine birds from different sampling sites. Error bars represent one standard error. An outlier with extremely high level of OHPs in Wuhan (WH) was excluded when data statistics.

than 2% in GZ and BJ. Similar geographic distributions (urban > nonurban) were previously showed in the terrestrial passerine studies from Europe and South China (Sun et al., 2012a; Van den Steen et al., 2009, 2010). PBDE concentrations in WH was significantly lower than that of the other two urban sites (Tukey HSD, $p < 0.01$), which was in accordance with the PBDE geographical profile showed in indoor dust samples from WH and GZ (Huang et al., 2010). However, it should be treated cautiously that species-specific accumulation may partly be responsible for the level variation among WH and the other sampling sites.

DBDPE was detected in 87% of urban samples but was non-detectable in the reference site. The geographic distribution of DBDPE in the three urban sites followed the order of GZ (median of 31 ng/g lw) > BJ (median of 8.5 ng/g lw) > WH (median of 3.6 ng/g lw), although no statistically difference was found ($F_{2,56} = 5.052$, $p = 0.065$, Fig. 2). As the replacement for Deca-BDE, DBDPE was extensively used in electrical and electronic industries (Covaci et al., 2011). Depositional records from the Dongjiang River (Zhang et al., 2009) suggested the recent source inputs of DBDPE in the Pearl River Delta where GZ located. Hu et al. (2010) reported the usage and potential source emission of DBDPE in surface sediments from North China where BJ was situated. Manufacturing of electronic equipment has become one of the pillar industries in GZ and BJ, constituting 12% and 16% of the gross output value of local industry in 2010, respectively (calculated based on the corresponding local statistical yearbook 2011). However, the gross industrial output value of electronic appliance manufacturing in WH was only one third or fourth of that in BJ or GZ. Therefore, emission from the regional industrial activities may contribute to the high level of DBDPE in GZ and BJ.

DP was detectable in all the sampling sites at concentrations ranging from non-detectable (nd) to 350 ng/g lw (Table 1). DP levels in GZ (median of 13 ng/g lw) were significantly higher than those in BJ (median of 4.9 ng/g lw) and WH (median of 4.4 ng/g lw) ($p < 0.01$, Fig. 2). DP has been used as a flame retardant in electrical hard plastic connectors in televisions and computer monitors, wire coatings, and so on for at least 40 years (Sverko et al., 2011). The relatively high DP concentrations in south of China compared to that in the north may be ascribed to the intensive e-waste recycling activities in southern region. Numerous studies have demonstrated the elevated DP levels in biotic and abiotic matrices from the e-waste recycling areas, South China (Sun et al., 2012b; Wang et al., 2011; Wu et al., 2010; Zheng et al., 2010). No significantly difference in DP level was found between GZ and the reference site (Tukey HSD, $p = 0.892$). In another biomonitoring study using passerine birds from South China, Sun et al. (2012b), either, did not observe significantly difference in DP levels between the rural/suburban sites and the urban sites.

No HBCDs were detected in the reference site. α-HBCD was the main isomer detected in birds. ΣHBCD concentrations ranged from 0.68 ng/g lw in GZ to 1100 ng/g lw in BJ in three urban regions (Table 1). Levels of HBCDs were significantly higher in BJ (median of 51 ng/g lw) than in the other two cities ($p < 0.001$, Fig. 2). Previous reports have demonstrated the elevated HBCD levels in ambient air in the north of China compared to that in the south (Hu et al., 2011; Yu et al., 2008), which was consistent with our results. The manufacturers of HBCDs in China are mainly located in the coastal areas of the eastern and northeastern China (Tanabe, 2008), which may contribute to the increasing HBCD levels in BJ.

3.3. Comparison with other areas

Data on OHPs in passerine birds are by far mainly concentrated in Europe and North America, of which DDTs, PCBs and PBDEs levels are mostly detected (Table S2 in Supplementary Data). Compared with the reports from other regions except for China, the PCB concentrations in the present study obviously lie at the low end of worldwide figures. The historically small volume of production and consumption on PCBs in China are probably responsible for this case (Xing et al., 2005). PBDE level in our study were in the same range as those of North America, but were generally higher than those of Europe (Chen et al., 2012; Eens et al., 2013) (Table S2 in Supplementary Data). These spatial distribution may mirror the uneven PBDE use patterns around the world, in which North America and Asia consumed 49% and 37% of the overall PBDE commercial formulation (including Penta-, Octa- and Deca-BDE), respectively, more than that consumed by the European market (12%) (Fig. S2) (Chen et al., 2012).

Presently, little information is available for non-PBDE FRs, such as DBDPE, DP and HBCDs, in terrestrial passerine birds. Levels of DBDPE and DP in the sparrows from GZ (median of 31 ng/g lw for DBDPE, and median of 13 ng/g lw for DP, respectively) were comparable with the previous reported concentration in passerine birds from the same region (median of 51 ng/g lw for DBDPE, and 23 ng/g lw for DP, respectively) (Sun et al., 2012a,b). DP concentrations in birds from GZ were also in the same order of magnitude as that in European starlings eggs from southern Lake Ontario region (0.2–10 ng/g wet weight with 5% lipid content) (Chen et al., 2012).

3.4. Spatial variation of contamination profiles

It is of importance to investigate contamination profiles for further concern on local contamination sources (Van den Steen et al., 2009). In the present study, significant spatial variations were demonstrated by the contaminant profiles among sampling sites. In the reference site, DDTs were generally the dominating contaminants, accounting for 51% in the total OHPs (Fig. 3), followed by equivalent PCBs, PBDEs and other non-PBDE FRs (19%, 15% and 15%, respectively). The dominance of DDTs in the total OHPs was consistent with that agriculture as the traditional activity in the reference site. Regarding the three cities, obviously dominating contribution of DDTs was found in WH whereas the predominant pollutants in BJ and GZ were replaced by PBDEs and other FRs (Fig. 3). The larger extent of agriculture in WH than that in BJ and GZ may attribute to the greater proportion of DDTs as discussed above. The dominance of PBDEs and other FRs in BJ and GZ showed

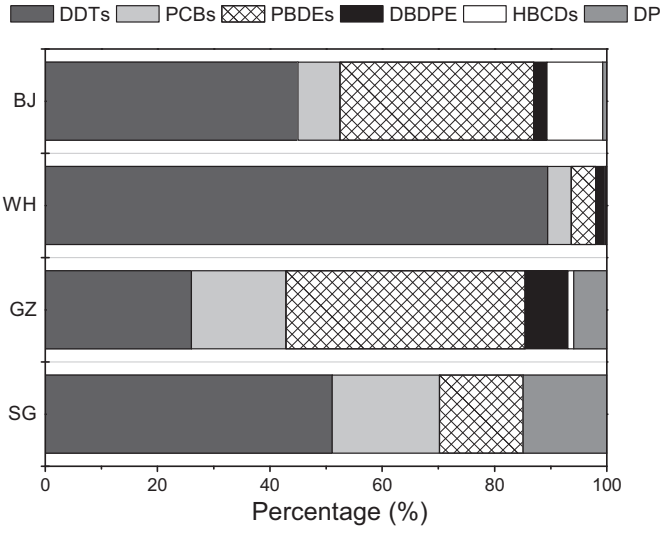

Fig. 3. Contamination profiles of investigated OHPs in passerine birds from different sampling sites.

discrepancy to most Chinese reports (Chen et al., 2009; Gao et al., 2009; Lam et al., 2008) in which DDTs were dominated. Elevated PBDE and other FR levels were previously linked to the point-source input such as e-waste recycling activities in the study area (Luo et al., 2009). In the present study, however, no e-waste recycling activities occurred in the sampling locations. The distribution pattern, thus, may mirror the integrate contaminants structure, of which industrial sources contributed more than agrochemical sources, in BJ and GZ. The transformation from traditional agriculture characterized contamination profile to industry characterized profile in some of the developed metropolises of China indicates that more attention should be attracted on the environmental risk brought about by the relevant pollutants such as FRs.

To gain more details on the source of PCBs and PBDEs in the study areas, congener profiles of PCBs and PBDEs were analyzed. CB-153, -138, -118, -180, and -28 were found to be the key congeners in all samples (Fig. S3). This was consistent with those in previous studies. However, PCB patterns showed significant differences among the four areas. WH samples showed more contributions of high chlorinated congeners, while SG samples exhibited more contributions of low chlorinated congeners than samples from BJ and GZ (Fig. 4), suggesting that the source of PCBs in different sampling sites could be different. A factor analysis based on PCB homologues composition revealed that samples from BJ overlapped a part of the samples from GZ, which implied they share the same PCB sources (Fig. S4). The samples from the university campus site (GZ2) overlapped the samples from SG (Fig. S4). When we look back upon the result of stable isotopic analysis, we found that these GZ2 samples share the same $\delta^{13}C$ values with samples from SG. This result indicated that diet source plays a key role in determination of PCB congener profile (Zimmermann et al., 1997).

PBDE congener profiles in birds from WH were significantly different from those in other three sites. The contribution of low brominated congeners (BDE-47, -100, -99, -154, -153 and -183) was significantly higher in WH than in other three sites (Fig. S5). This was likely caused by the difference in consumption of PBDE technical mixtures among different regions. The relatively amount of Penta- and Octa-BDE mixtures used in WH may be higher than in other regions. Alternatively, species-specific difference in metabolism of PBDEs could also contribute to this PBDE congener profile difference.

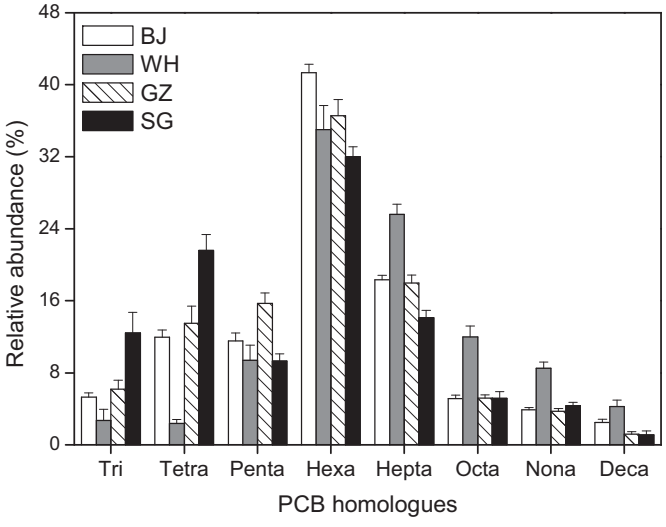

Fig. 4. PCB homologue profiles in passerine birds from different sampling sites.

4. Conclusions

Our results revealed geographical variation in the levels and contamination profile of OHPs in passerine birds from three metropolises of China, which generally reflected the emission patterns in the corresponding sampling site. Specifically, Beijing and Guangzhou with higher degree of industrialization and urbanization showed elevated contribution of FRs. Differential $\delta^{13}C$ values, referring to different diet sources, may contribute to the PCB homologues composition variation. Relatively higher $\delta^{13}C$ values were found in urban birds compared to those from the non-urban sites, indicating the anthropogenic effects on urban ecosystems. Given that sparrows are human commensals and are ubiquitous residential birds along rural–urban environmental gradients, it may be feasible to use this species as a biomonitoring tool on a large geographical scale for further investigation in contamination linked to human activities.

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Appendix A. Supplementary data

Supplementary data related to this article can be found at http://dx.doi.org/10.1016/j.envpol.2014.01.023.

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