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A novel pollution pattern: Highly chlorinated biphenyls retained in Black-crowned night heron *(Nycticorax nycticorax)* and Whiskered tern *(Chlidonias hybrida)* from the Yangtze River Delta

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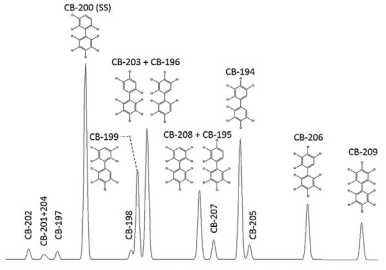
HIGHLIGHTS

- A number of POPs were determined in two water bird eggs from Yangtze River Delta.
- Highly chlorinated biphenyls (CB194-209) were detected in all individual eggs.
- 4,4′-DDE was the predominant pesticide in all samples, followed by β-HCH and Mirex.
- The exposure level of POPs was in the median or low level in the world.

GRAPHICAL ABSTRACT



Highly chlorinated biphenyls (8-10 chlorine substituents) chromatography pattern in bird egg

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ABSTRACT

Contamination of organochlorine pesticides (OCPs), polychlorinated diphenyls (PCBs), polybrominated diphenyl ethers (PBDEs), hydroxylated polybrominated diphenyl ethers (OH-PBDEs) and their methylated counterparts (MeO-PBDEs) were determined in Black-crowned night heron *(Nycticorax nycticorax)* and Whiskered tern *(Chlidonias hybrida)* from two drinking water sources, e.g. Tianmu lake and East Tai lake in Yangtze River Delta, China. A novel PCBs contamination pattern was detected, including 11% and 6.9% highly chlorinated biphenyls (PCBs with eight to ten chlorines) in relation to total PCB concentrations in the Black-crowned night heron and Whiskered tern eggs, respectively. The predominating OCPs detected in the present study were 4,4′-DDE, with concentration range 280–650 ng g^{-1} lw in Black-crowned night heron and 240–480 ng g^{-1} lw in Whiskered tern, followed by β-HCH and Mirex. 6-MeO-BDE-90 and 6-MeO-BDE-99 are the two predominant congeners of MeO-PBDEs whereas 6-OH-BDE-47 contributes mostly to the OH-PBDEs in both species. Contamination level was considered as median or low level compared global data.

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1. Introduction

The Yangtze River Delta (YRD) is well-known as the "land of fish and rice", as it is made up of a large number of lakes and rivers, and suitable for rice growing and freshwater fish. The Yangtze River basin provides more than 70% of the country's rice production and 50% of grain production, accounting for 40% of China's gross domestic product (Floehr et al., 2013). Both Tianmu lake (TML) (Li et al., 2012) and East Tai lake (ETL) (Tao et al., 2010; Yu et al., 2013) are important drinking water sources in YRD. However, Yu and coauthors (Yu et al., 2013) found that the chloride concentration increased 6.76 folds in Tai lake from 1950s to 2010s, and its current water chemistry has become "anthropogenic dominance" from its original rock dominance. The current concentrations of hexachlorocyclohexanes (HCHs), heptachlor, heptachlor epoxide, aldrin, dieldrin in the shallow groundwater in majority of the areas of Taihu Lake region may pose serious cancer risk to the local population, especially to children (Wu et al., 2014).

Large numbers of scientific studies have reported on the occurrence and effects of persistent and bioaccumulative organic pollutants in wildlife, humans and abiotic matrices over the last 50 years. Some of these, e.g. twenty-three persistent organic pollutants (POPs) or classes of chemicals are regulated by the Stockholm Convention (UNEP, 2015). The largest group of POPs is organochlorine pesticides (OCPs), which have a long history of production (IPEP, 2006) and use around the world, not the least dichlorodiphenyltrichloroethane (DDT) in China. Other important groups of POPs, commercially produced in large quantities, are the industrial chemical, polychlorinated biphenyls (PCBs) and polybrominated diphenyl ethers (PBDEs). Only concentrating on some selected recent reports on these pollutants are reported in soil (Zhang et al., 2009), sediment (Mai et al., 2005), mussels (Yin et al., 2015), fish (Qiu et al., 2012) and birds (Dong et al., 2004; Nakata et al., 2005) from China. In addition to the POPs, some natural product and/or metabolites of PBDEs are of emerging concern due to their occurrence in wildlife, e.g. hydroxylated polybrominated diphenyl ethers (OH-PBDEs) and methoxylated polybrominated diphenyl ethers (MeO-PBDEs) (Malmvarn et al., 2005; Lofstrand, 2011; Dahlberg et al., 2014).

Bird and bird eggs have been successfully used as POPs monitoring species as they are widely spread, sensitive to environmental changes from anthropogenic sources and in high trophic level in the food chain (Jaspers et al., 2006; Voorspoels et al., 2006; Park et al., 2009; Sun et al., 2012). Previous studies have been conducted on the POPs in avian species in Europe as e.g. continuously reported by the Swedish EPA (Swedish EPA, 2013) and in individual species (Jorundsdottir et al., 2010, 2013; Gomez-Ramirez et al., 2014). In China, several studies on POPs in bird species have been conducted in recent years. However, most of the studies have been focused on the Pearl River Delta (Luo et al., 2009; Liu et al., 2010; Sun et al., 2012). Although YRD is one of the most developed areas in China, the scientific data regarding POPs on birds is very limited.

Whiskered tern (*Chlidonias hybrida*, WT) and Black-crowned night heron (*Nycticorax nycticorax*, NH) are common species in East Tai lake and in Tianmu lake of the YRD, respectively. Both of them feed mainly on small fish, amphibians, crustaceans, insects and their larvae. However, study on wetland bird diversity in Anqing floodplain wetlands, middle-lower reaches of the Yangtze River showed that Whiskered tern is the dominant species in both water habitats and reed marsh habitats, while Black-crowned night heron are abundant in both water habitats and farmland habitats (Gong et al., 2013), indicating a little difference between two species.

The objective of the present study was to determine concentrations and patterns of OCPs, PCBs, PBDEs and OH-/MeO-PBDEs in bird species from YRD. Hence, eggs of the two bird species indicated above were collected from two drinking water sources sites (ETL and TML) in the YRD, China.

2. Materials and methods

2.1. Samples and sampling

Ten Black crowned night heron (*N. nycticorax*, NH) eggs were collected from Tianmu lake (TML) and ten Whiskered tern (*C. hybrida*, WT) eggs from East Tai lake (ETL) (Fig. 1). The eggs were collected in April and May, 2014. All eggs were collected randomly without any conscious selective criterion with the help of local fishermen. The eggs were immediately sent to the laboratory. The egg surface was cleaned with deionized water, and the egg white and yolk was blown out and homogenized prior to storage at −20 °C, until taken out for chemical analysis.

2.2. Chemicals and standards

All solvents used were of pesticide quality. Authentic reference standards of OCPs, including 4,4′-DDT, 4,4′-DDE, 4,4′-DDD, 2,4′-DDT, 2,4′-DDE and 2,4′-DDD, α-HCH, β-HCH, γ −HCH were purchased as a mixture from Larodan Fine Chemicals (Malmö, Sweden). The PCB congeners including CB-28, 52, 101, 105, 118, 128, 138, 146, 153, 156, 170, 180, 183, 187 and 189 were purchased from Larodan Fine Chemicals (Malmö, Sweden). The highly chlorinated biphenyls with at least eight chlorines (PCBs(Cl_{8-10})): CB-194 − CB-209 were purchased from AccuStandard (New Haven, USA). PBDE congeners: BDE-28, 47, 66, 99, 100, 153, 154 and 183 were purchased from LGC Promochem (Wesel, Germany). MeO-PBDEs congeners, including 2′-MeO-BDE-28, 6-MeO-BDE-47, 2′-MeO-BDE-68, 6-MeO-BDE-90 and 6-MeO-BDE-99 were synthesized in-house (Marsh et al., 2003, 2005).

Gel permeation chromatography (GPC) was performed on Bio-Beads SX-3 gel (200–400 mesh size) from Bio-Rad laboratories (CA, USA). Diazomethane was prepared in-house from *N*-methyl-*N*-nitroso-*p*-toluenesulfonamide (Sigma–Aldrich, Steinheim, Germany). Working with diazomethane has been approved by the Swedish work authority (IMS 2012/39924).

2.3. Extraction and clean up

Four gram of egg white and yolk was homogenized. Prior to extraction, surrogate standards (SS): CB-200 (5.2 ng), BDE-139 (3.4 ng), Dec 603 (7.2 ng) and 4′–OH–BDE-121 (0.9 g) were added to the samples. The extraction was performed according to Jensen et al. (Jensen et al., 2009), except that *iso*-hexane (*iso*-hxn) replaced *normal*-hexane (*n*-hxn). After extraction, the lipid content of each sample was determined gravimetrically. GPC was applied to remove lipids because of the high lipid content. The GPC was equipped with an injection loop (1 mL) and a glass column (500 × 25 mm i.d.) packed with Bio-Beads SX-3 gel. *Iso*-hxn: Dichloromethane (1:1, v/v) with 0.5% formic acid was used as the mobile phase. The flow rate was set to 4 mL/min. The samples were injected to GPC and divided into two fractions. The first fraction 0–32 min containing lipid together with chlorinated paraffins was saved for future analysis. The second fraction from 32 to 60 min, containing the analytes of interest for the present study was collected. The samples volume was adjusted to 4 mL prior to potassium hydroxide partitioning (0.5 M in 50% ethanol) to separate phenolic compounds from neutral compounds (Jensen et al., 2009). The isolated phenolic compounds were derivatized with diazomethane in excess amount with great care and further analyzed for

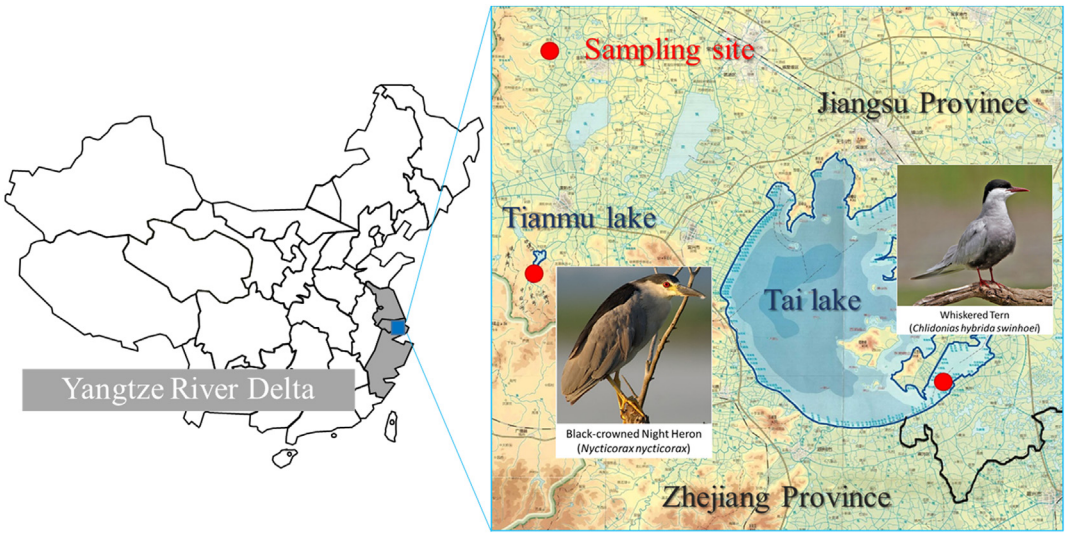

Fig. 1. Sampling sites in the Yangtze River delta for the eggs of Black-crowned night heron (*Nycticorax nycticorax*, NH) and Whiskered tern (*Chlidonias hybrida*, WT).

brominated substances. Pentachlorophenol was quantified but not included in the results. The methylation reaction lasted for 3 h at room temperature in darkness. Prior to instrumental analysis, 6-MeO-BDE-137 (4.0 ng) and ϕ-DDE (6.0 ng) were added into the samples as volumetric standard (VS). The results were present on lipid weight concentration (ng g^{-1} lw).

2.4. Instrumental analysis

OCPs and $\sum_7$PCBs were analyzed by Varian 450 gas chromatogram equipped with electron capture detector (GC-ECD) and Varian CP-8400 auto-sampler. The injector (1 μL) was operated in the programmable temperature vaporizing (PTV) mode at temperature of 260 °C. The column was a BP5 (30 m × 0.25 mm i.d. × 0.25 μm film thickness; SGE Analytical Science) with helium as a carrier gas and nitrogen as make-up gas. The column oven temperature program was 80 °C for 2 min, 15 °C/min to 300 °C and hold for 18 min.

PBDEs and MeO-PBDEs were analyzed by Varian-450 gas chromatogram coupled to a Varian 320 mass spectrometry (GC–MS) using electron capture negative ionization (ECNI) and selective ion monitoring SIM) mode, scanning bromine ions (*m*/*z* 79 and 81). Automated 1 μL injections with a CTC GC Pal auto sampler was conducted on a DB-5HT(30 m × 0.25 mm i.d. × 0.10 μm film thickness; Agilent J&W) GC column, with methane (scientific 5.5, AGA Stockholm, Sweden) as reagent gas. The injector was operated in PTV mode at temperature of 260 °C. Helium was used as carrier gas at a set constant flow of 1.0 mL/min. The oven program was 55 °C for 2 min, 15 °C/min to 320 °C and hold for 4 min. The ion source and transfer line temperature were set at 230 °C and 300 °C, respectively.

$Cl_{8\text{-}10}$PCBs were analyzed with the same instrumental condition as PBDEs and MeO-PBDEs, except that the scanning ions were 427.7/429.7, 461.6/463.6, 497.7/499.7 and 635.7/637.7 for octa-CBs, nona-CBs and deca-CB and Dec 603 (SS), respectively. All of the identification and quantification are based on authentic reference standards.

2.5. Quality assurance and quality control

One procedural solvent blank was analyzed in parallel with each batch of five samples to assess any potential contamination during laboratory work. Limit of detection (LOD) was set to three times the background noise (S/N = 3). Limit of quantification (LOQ) was set to 10 times (S/N = 10) the background noise or three times the level in the procedural solvent blank if it was detected. BDE-47, - 99 and 153 were present in blanks and were subtracted from the samples. The range of recoveries (mean ± standard deviation(S.D.) of SS were 92–111% (100% ± 5%), 73–112% (79% ± 7%) and 74–130% (84% ± 13%) for CB-200, Dec 603 and BDE-139, respectively. Low recoveries were obtained for 4′–OH–BDE-121 in the present study.

2.6. Statistics

Mann-kendalls tau test was applied to examine a potential correlation between PBDEs and corresponding OH-/MeO-PCDEs. Principal component analysis (PCA) was conducted on the fractional composition of PCBs by Unscrambler 10.3 to evaluate the sources of PCBs in bird eggs compared with technical Aroclor products.

3. Results

The mean and median concentrations on lipid weight of PCBs, OCPs, PBDEs, OH-PBDEs and MeO-PBDEs, in the NH and WT, are reported in Table 1. CB-153 was the most abundant PCB congener in both species, followed by CB-138 and CB-118. CB-209 was detected in all bird eggs with the mean concentration of 6.4 and 4.2 ng g^{-1} lw in NH and WT eggs, respectively. In total nine individual PCB congeners with eight or more chlorines are quantified and as potential mixtures another six congeners. The congener patterns of PCBs assessed in the NH and WT are shown in Fig. 2. The highly chlorinated PCBs ($Cl_{8\text{-}10}$) accounted for 11% and 6.9% of the $\sum$PCB amount in NH and WT, respectively. The concentrations of highly chlorinated biphenyls, PCBs ($Cl_{8\text{-}10}$), excluding CB-200, ranged 20–100 and 11–150 ng g^{-1} lw with the mean concentration of 47 and 38 ng g^{-1} lw, in NH and WT, respectively. The concentrations of the PCBs ($Cl_{8\text{-}10}$) are presented in Table 1.

4,4′-DDE was the dominating OCPs in the two bird species eggs, with the mean concentration of 450 and 320 ng g^{-1} lw in NH and WT, respectively. β-HCH was the most abundant HCH isomer in all samples analyzed. Mirex was detected in all samples with the mean concentration of 94 and 41 ng g^{-1} lw in NH and WT, respectively.

Of all the 8 PBDE congeners analyzed, BDE-28 and -66 were not detected in any NH while BDE-183 was detected in only five WT

Table 1
Concentration (ng g^{-1} lw) of some prioritized polychlorinated and polybrominated organic pollutants in Black-crowned night heron (*Nycticorax nycticorax*, NH) and Whiskered tern (*Chlidonias hybrida*, WT) from Tianmu lake (TML) and East Tai lake (ETL), respectively.

Compounds	Black-crowned night heron (*Nycticorax nycticorax*, n = 10)				Whiskered tern (*Chlidonias hybrida*, n = 10)			
	Mean	Median	Range	S.D.	Mean	Median	Range	S.D.
4,4′-DDE	450	460	280–650	120	320	290	240–480	72
4,4′-DDD	41	24	2.4–150	48	4.2	3.3	1.3–11	3.6
4,4′-DDT	18	6.1	1.8–110	32	18	2.7	2.0–90	30
∑DDTs[a]	520	530	290–780	180	360	340	270–610	99
β-HCH	300	190	150–850	250	79	56	30–270	69
∑HCHs[b]	300	190	150–850	250	80	56	30–270	70
Mirex	94	49	17–340	110	41	28	9.3–160	44
CB-28	8.4	5.1	2.2–29	8.6	22	11	3.9–120	34
CB-101	9.3	5.7	3.6–29	7.8	35	9.3	4.0–140	55
CB-118	57	43	20–180	45	71	25	9.9–230	85
CB-138	61	43	17–150	44	71	28	14–220	78
CB-153	110	74	39–280	79	120	59	32–330	120
CB-180	60	29	7.7–110	35	41	9.2	4.2–190	65
CB-194	7.4	7.2	2.4–14	4.2	6.0	2.4	1.2–23	7.3
CB-197	2.8	1.8	1.4–11	2.9	1.7	0.82	0.52–5.1	1.6
CB-198	0.59	0.49	0.37–1.5	0.31	0.5	0.28	0.16–1.8	0.52
CB-199	6.3	5.5	2.4–15	4.2	7.9	2.7	1.4–42	12
CB-201 + 204	1.6	1.2	0.73–5.7	1.5	1.8	0.77	0.52–5.9	1.9
CB-202	1.1	1.0	0.38–2.2	0.57	0.91	0.36	<LOQ-4.7[h]	1.4
CB-203 + 196	8.2	7.7	2.8–16	4.6	7.2	2.5	1.2–36	11
CB-205	1.3	0.91	0.42–3.9	0.99	0.48	0.49	0.16–0.88	0.29
CB-206	4.3	3.7	1.9–11	2.6	2.1	1.1	0.68–5.6	1.7
CB-207	2.3	1.8	1.3–5.9	1.4	1.3	0.87	0.48–3.6	1.0
CB-208 + 195	4.6	4.1	1.8–9.7	2.7	3.8	1.3	0.62–19	5.7
CB-209	6.4	5.6	3.5–16	3.6	4.3	3.8	1.3–15	4.0
∑PCBs (Cl_{8-10})[c]	47	44	21–103	25	38	16	11–148	44
$\sum_{30}$PCBs[d]	410	330	150–980	260	550	190	120–2100	680
BDE-47	4.7	3.9	1.2–15	4.0	45	11	6.8–260	79
BDE-99	3.7	1.7	0.65–19	5.5	24	5.0	2.1–200	61
BDE-100	6.4	3.9	2.0–29	8.0	11	4.2	2.3–56	16
BDE-153	11	6.7	4.6–41	12	6.0	2.3	0.88–29	8.9
BDE-154	15	10	6.7–47	12	7.5	3.9	2.3–21	6.9
BDE-183	2.1	1.9	0.76–5.2	1.3	0.83	0.40	<LOQ-4.1[i]	1.3
$\sum_8$PBDEs[e]	43	29	19–150	41	99	28	15–580	170
6-MeO-BDE-47	1.3	0.21	<LOQ-10	3.1	0.57	0.56	<LOQ-1.4[j]	0.35
6-MeO-BDE-99	3.3	1.5	0.40–20	5.8	2.1	1.8	0.98–4.9	1.1
$\sum_5$ MeO-PBDEs[f]	8.4	3.7	0.86–50	15	5.4	4.4	3.2–12	2.5
6-HO-BDE-47	3.4	0.97	0.16–12	4.1	2.0	1.4	0.16–5.8	1.6
$\sum_5$ HO-PBDEs[g]	3.5	1.1	0.16–12	4.2	2.0	1.4	0.16–5.9	1.7

[a] Sum of 2,4′-DDE, 2,4′-DDD, 2,4′-DDT, 4,4′-DDE, 4,4′-DDD and 4,4′-DDT.
[b] Sum of α-HCH, β-HCH and γ-HCH.
[c] Sum of CB-194–209 (except for 200).
[d] Sum of CB-28, 52, 101, 105, 118, 128, 138, 146, 153, 156, 170, 180, 183, 187, 189 and 194–209 (except for 200).
[e] Sum of BDE-28, 47, 66, 99, 100, 153, 154 and 183.
[f] Sum of 2′–OH–BDE-28, 6-OH-BDE-47, 2-OH-BDE-68, 6-OH-BDE-90 and 6-OH-BDE-99.
[g] Sum of 2′-MeO-BDE-28, 6-MeO-BDE-47, 2-MeO-BDE-68, 6-MeO-BDE-90 and 6-MeO-BDE-99.
[h] LOQ for CB-202 was 0.05 ng.
[i] LOQ for BDE-183 was 0.04 ng.
[j] LOQ for 6-MeO-BDE-47 was 0.08 ng.

samples. Mean concentrations of $\sum_8$PBDEs were 43 and 99 ng g^{-1} lw in NH and WT, respectively. 6-MeO-BDE-90 and 6-MeO-BDE-99 are the two predominant congeners of MeO-PBDEs in both species while 6-OH-BDE-47 contributes mostly to the OH-PBDEs (cf. Table 1 and Fig. 2).

4. Discussion

4.1. PCBs

The high abundance and pattern of the highly chlorinated biphenyls reported herein (Table 1 and Fig. 2) are novel and remarkable. Except CB-202, with 95% of detected frequency, the other Cl_{8-10}PCBs congeners were detected in all bird eggs. The high contribution of PCBs (Cl_{8-10}) to total PCBs indicated a specific but yet unknown source of PCBs in YRD, China. A previous study reported CB-209 in surface sediment in upstream of Yangtze River (Yang et al., 2009) and also in some aquatic species from e-waste recycling site in South China (Wu et al., 2008). CB-209 was detected as the most abundant congener in fish from Liaohe river in Northeast China, contributing 44% to ∑PCBs (Ren et al., 2013). To our knowledge, this is the first report on high abundance and relatively high concentrations of PCB congeners with all of eight to ten chlorine substituents (CB-194 to CB-209) in wildlife in general, and indeed in bird eggs from the YRD. Kannan et al. (Kannan et al., 1997) found octa-CBs and nona-CBs in soil and sediment from a site in Georgia. They suggested a contamination with Aroclor 1268 (85% Cl_{8-10}), which is a higher chlorinated technical mixture than Aroclor 1260 (8.5% Cl_{8-10}) could be the source. In the present study, CB-199 and CB-203/196 was the predominant CB congeners (Cl_{8-10}) in WT with concentration of 7.9 ng g^{-1} lw and 8.2 ng g^{-1} lw, respectively. A descending concentration trend of congeners with CB-199, 208/205,194 and 209 was observed in WT, whereas a slightly difference order of CB-203/196, 194,209 and 194 was found

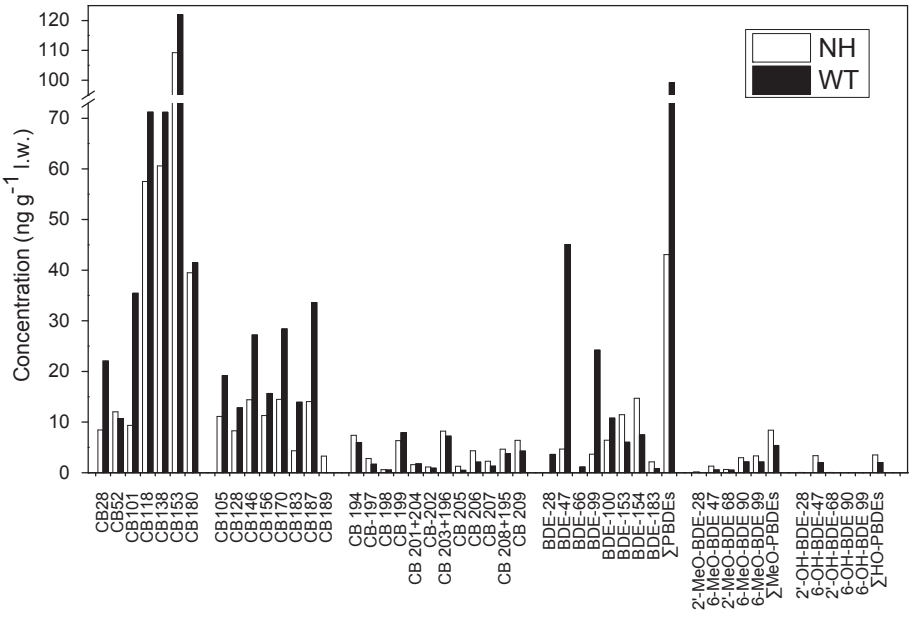

Fig. 2. Concentrations of PCBs, including highly chlorinated biphenyls, PBDEs, MeO-PBDEs and OH-PBDEs in Black-crowned night heron (*Nycticorax nycticorax*, NH) and Whiskered tern (*Chlidonias hybrida*, WT) from Yangtze River Delta.

in NH (Table 1). The ratio between CB-209 and CB-207 was 2.1–4.3 and 1.7–6.6 in NH and WT, respectively, which is one magnitude higher than in commercial Aroclor 1262 (0.22) but close to that of Aroclor 1268 (3.3) (Anderson, 1991). However, Howell et al. (2008) suggested that other sources than Aroclor might influence the PCBs pattern (Howell et al., 2008). For instance, Hu and coauthors (Hu et al., 2011) analyzed sediment cores from Lake Ontario and Indiana Harbor Ship Center and found that CB-209, CB-208, CB-207 and CB-206 reached a peak in the 1950s. They (Hu and Hornbuckle, 2010) have pointed out that deca-CB and nona-CBs were present in phthalocyanine green pigment. Further, CB-209 is also suspected to be formed during the process pf titanium dioxide purification (Rowe et al., 2007). The occurrence of PCBs ($Cl_{8\text{-}10}$) in both species collected from two sites in YRD together with the considerable concentrations reported need to be further studied, not the least potential sources requires identification. The present report also calls for more congeners specific studies on occurrence and toxicity of these congeners.

The concentrations of $\sum_7$ PCBs were in the range of 88–730 and 86–1200 ng g^{-1} lw in NH and WT eggs, respectively. The concentration of $\sum_7$ PCBs was in accordance with those in passerine birds in Pearl River Delta (Yu et al., 2014) and in common tern from Shandong province (Gao et al., 2009) but much lower than those in Caspian tern and Forster's tern from San Francisco Bay, USA (She et al., 2008), which indicated PCBs contamination from traditional technical production was less severe in China. The CB-153 concentrations in the NH and WT eggs seem to be slightly lower than in Latvian Grey heron (*Ardea cinerea*) chicken blood (Valters, 2001). However, $\sum_7$ PCBs in the NH and WT eggs were much higher than mandarin fish (27 ng g^{-1} lw) (Qiu et al., 2012) and blue mussels (15 ng g^{-1} lw) (Yin et al., 2015) in the same region confirming bioaccumulation and biomagnification of PCB congeners in the aquatic ecosystem.

The PCA shown in Fig. 3 was conducted on the fractional composition of PCBs in birds eggs compared with technical Aroclor products to ascertain whether multiple sources were contributing to the PCBs or not. The first two principal components accounted for 37% and 32% of the total variance, respectively. It showed that the pattern of PCBs found in bird eggs in the present study was similar to Aroclor 1254 and 1260, which was consistent with the common thought that the source of PCBs in China is corresponding to the industrial product like Aroclor 1254 (Yu et al., 2005). However, most of the samples got the positive score for the first principal component, which resulted from both the predominant PCB congeners (CB-153 and 138) and the PCBs ($Cl_{8\text{-}10}$). Considering the level of PCBs ($Cl_{8\text{-}10}$) could not be overlooked, the PCBs pattern found in the present study were not only from historic usage but also from present sources. It could be either from a higher chlorinated PCB mixture similar to Aroclor 1268 and 1270 or other sources such as phthalocyanine green pigments and/or titanium dioxide purification.

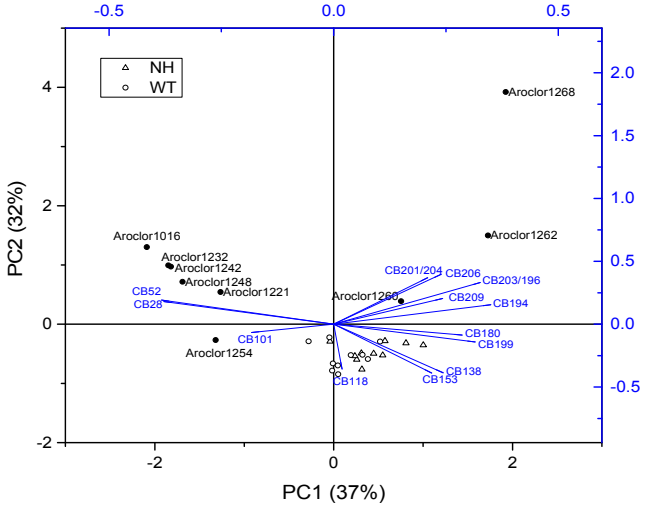

Fig. 3. Principal component analysis on seven PCB congeners and highly chlorinated biphenyls in Black-crowned night heron (*Nycticorax nycticorax*, NH), Whiskered tern (*Chlidonias hybrida*, WT) and Aroclor commercial mixtures from Yangtze River Delta.

Table 2
Comparison of PCBs, OCPs and PBDEs level (ng g^{-1} l.w.) in Black-crowned Night Heron (*Nycticorax nycticorax*, NH) and Whiskered Tern (*Chlidonias hybrida*, WT) in the present study with some other Chinese and international studies.

Species name	Position	year	n	CB-153	CB-138	∑PCBs	4,4′-DDE	∑DDTs	∑HCHs	Mirex	BDE-47	BDE-99	BDE-153	∑PBDEs	Reference
Night heron	Liyang, China	2014	10	110	61	280	450	500	300	94	4.7	3.7	11	(25–150)	Present study
Night heron	Xiamen, China	2004	5				31000	40000		320	29	7.5	19	120	(Lam et al., 2007; Lam et al., 2008)
Night heron	Quanzhou, China	2004	5				32000	41000		145					(Lam et al., 2008)
Night heron	Wuxi, China	2008	65				5500[a]	5600[a]	460[a]						(Dong et al., 2004)
Night heron	Hongkong, China	2006	16				2300	2500		84					(Wang et al., 2011)
Night heron	Hongkong, China	2004	5								110	38	56	400	(Wang et al., 2012)
Night heron	Salton Sea	2004	11				45	62	NC[b]	4.3					(Henny et al., 2008)
Night heron	Wisconsin, USA	2010	4				410[c]	410[c]		1.5[c]					(Custer and Custer, 1995; Custer et al., 2014)
		1991	7				2100[c]								
Chinese pond heron	Wuxi, China	2000	43				2700[a]	2800[a]	280[a]						(Dong et al., 2004)
Chinese pond heron	Hongkong, China	2006	12				4320	4760		130					(Wang et al., 2011)
Great blue heron	Fraser River, USA	2012	5								27	16	8.9	72	(Miller et al., 2015)
Whiskered tern	Suzhou, China	2014	10	120	71	360	320	340	80	41	45	24	6.0	(15–580)	Present study
Caspian tern	San Francisco Bay, USA	2000	5	3000							1300	520	110	2600	(She et al., 2008)
Forster's tern	San Francisco Bay, USA	2000	5	2000							1000	670	170	2200	(She et al., 2008)
Common tern	Shandong, China	2000	9			130–660		1400–2000						34–230	(Gao et al., 2009)
Common tern	Netherlands	2008	5								90	35	17	200	(Brandsma et al., 2015)
Sooty Tern	Republic of Mauritius	2008	10	4.3	2.0	12	21	23	1.6	5.0					(Bouwman et al., 2012)
Arctic tern	Iceland	2003	6	160	130		280				73	11	3.8	110	(Jorundsdottir et al., 2010, 2013)

[a] Reported on ng g^{-1} dry weight.
[b] NC = no mean calculated, found in <50% of samples.
[c] Reported on ng g^{-1} wet weight.

4.2. OCPs

4.2.1. DDTs

Concentrations of DDTs were much higher than concentration of any other OCPs in present study. 4,4′-DDE derived from DDT accounted for 49% and 28% of total OCPs in NH and WT, respectively. It attained the highest concentration of total OCPs and detected in all samples. As we know, the use of technical DDT has been banned for agricultural purposes since over 30 years and the newly input into the environment can possibly be attributed to production of dicofol (Qiu et al., 2005). However, the low ratio (0.51 for NH and 0.07 for WT) between 2,4′-DDT and 4,4′-DDT indicated dicofol use might be not the main source for the DDT contamination in this area. The ratio between 4,4′-DDT and ∑DDTs (∑4,4′-DDT+4,4′-DDE+4,4′-DDD) was 0.029 and 0.034 in NH and WT, respectively. This ratio indicated that DDTs detected in the present study were mainly from historic usage. The ratio between 4,4′-DDT and 4,4′-DDTs in NH was consistent with other studies on DDTs in birds (Wang et al., 2011; Custer et al., 2014). The ratios are much lower than those for mussels (0.42–0.47) (Yin et al., 2015) and fish (0.38) (Qiu et al., 2012) in YRD. This difference in the ratio may be explained by the different metabolism rate in the species at different trophic level.

The concentration of DDTs found in our study was lower than reported from other parts of China (Lam et al., 2008; Wang et al., 2011), but comparable to heron from heron chicken blood from Latvia (Valters, 2001) and Arctic tern from Iceland (Jorundsdottir et al., 2010). However, the 4,4′-DDE level in heron was one magnitude lower than reported in another study (Dong et al., 2004) conducted in Taihu basin even though the HCHs levels were comparable. The contamination degree in the herons/heron eggs may be significantly influenced by hatching and feeding areas of the herons.

4.2.2. HCHs

β-HCH was the predominant HCH isomer in all samples. The concentration of ∑HCHs in NH was about four times higher than those in WT. It is difficult to explain what is causing this difference, e.g. differences due to species (food, uptake, metabolism) or differences between the contaminant burden between the two sites. Interestingly, β-HCH corresponded to almost 100% of ∑HCHs in the present study, which was slightly different from other biological matrices in previous studies from the YRD (Qiu et al., 2012; Yin et al., 2015) in which β-HCH was the predominant isomers but α-HCH and γ-HCH also detected. However, this pattern distribution was consistent with bird egg studies from other researches (Dong et al., 2004; Lundstedt-Enkel et al., 2005; Braune et al., 2007), indicating the species-specific difference of HCH isomer distribution in birds from other matrices. The ∑HCHs and HCHs isomer pattern distribution in NH in the present study was in the medium of global level and it was consistent with previous study on Tai lake (Dong et al., 2004) (cf. Table 2). Comparing β-HCH concentrations in NH and WT with blood concentrations in grey heron chickens from Latvia (Valters, 2001), the levels are clearly higher in the YRD species reported on herein.

4.2.3. Mirex

Mirex is another important OCP contaminant, together with 4,4′-DDE and HCH found in the present study (Table 1). As HCHs, the Mirex level in NH was higher than that in WT. Our results showed comparable level with bird eggs from North American (Champoux et al., 2006) and South China (Lam et al., 2008; Wang et al., 2011). Even though the Mirex level was comparable, they may be explained by different sources. Mirex was used as flame retardants and also pesticides in North American before 1980s,

however, it still can be detected with such level. In China, Mirex was produced until 2009 for control of termites and the estimated annually usage amount was about 300 kg (Yu et al., 2005).

4.3. PBDEs and related compounds

4.3.1. PBDEs

The mean concentration of ∑PBDEs in WT from ET lake was twice the levels in NH from TM lake, industrialization too small difference to enable us to speculate on the reason for this difference. The PBDEs concentrations found in the present study was in the median or low range, worldwide (cf. Table 2). BDE-47 and BDE-154 were the most abundant congeners in both WT and NH. PBDEs pattern profiles (Fig. 2) showed difference between the two species. The contribution of tri-, tetra-, and penta-BDEs (BDE-28, 47, 66, 99, 100) was greater in WT whereas hexa- and hepta-BDEs (BDE-153, 154 and 183) showed the opposite way. There may be several reasons for this difference including kinetics. BDE-209 was analyzed for but not detected as one of the main PBDE congeners in the present study which was not in accordance with other studies in Pearl river delta (Yu et al., 2014). This can be due to the levels of contamination in the area compare to other locations in China and outside.

4.3.2. OH-PBDEs

The OH-PBDEs congener profile in NH and WT was dominated by 6-OH-BDE-47, which composed 96% and 100% of the ∑OH-PBDEs. 6-OH-PBDEs level in our study was one order lower than those reported in black-head gulls (4.6 ng g^{-1} wet weight) from Bohai Sea, northern China (Zhang et al., 2012). 6-OH-BDE-47 is primarily produced naturally, but also a metabolite of BDE-47 in pike (Kierkegaard et al., 2004). However, the metabolism of BDE-47 in these bird species is unknown. No significant correlation were found between BDE-47 and 6-OH-BDE-47 ($p = 0.066$). The ratios of 6-OH-BDE-47/BDE-47 were 0.72 and 0.05 in NH and WT, respectively. The relatively high ratio in NH indicated that naturally production in algae as the main source of 6-OH-BDE-47 for NH. However, the ratio in WT is much smaller than in NH, and thus it is more difficult to assess the importance of the anthropogenic versus the natural product pathways.

4.3.3. MeO-PBDEs

In contrast to PBDEs, MeO-PBDEs showed higher level in NH than that in WT. MeO-PBDEs have been considered to be mainly naturally produced in the marine environment (Lofstrand, 2011), except that Feng et al. reported MeO-PBDEs in rainbow trout after exposure to decabromodiphenyl ether (Feng et al., 2010). In the present study, 6-MeO-BDE-90 and 6-MeO-BDE-99 were identified in all samples while 6-MeO-BDE-47 and 2′-MeO-BDE-68 were only detected in 70% and 65% of the samples. MeO-PBDEs congener profile differed from other studies in YRD (Qiu et al., 2012), which showed 6-MeO-BDE-47 and 2′-MeO-BDE-68 were the main congeners in fish. The relatively high percentage of 6-MeO-BDE-90 and 6-MeO-BDE-99 indicate a potential difference in the metabolism, as well as take-up ability between marine species and birds. Lofstand and coauthor (Lofstrand et al., 2011) suggested 6-MeO-BDE-90 and 6-MeO-BDE-99 could be formed by debromination of 6-MeO-BDE-137 in fish. No significant correlation was found between BDE-99 and 6-MeO-BDE-99 ($p = 0.38$), which could further support that MeO-PBDEs originate naturally rather than from anthropogenic source as metabolites of PBDEs. Even though low concentration of OH-PBDEs and MeO-PBDEs was detected in the present study, it might still be of some concern because these substances have association with endocrine disrupting effects and disruption of oxidative phosphorylation (Legradi et al., 2014).

5. Conclusion

The present study stresses a novel PCB contamination pattern, including 11% and 6.9% of PCBs ($Cl_{8\text{-}10}$) relative of total PCB content in the NH and WT, respectively. This finding implies further studies to identify sources of these highly chlorinated biphenyls. 4,4′-DDE, β-HCH and Mirex were the prevalent OCPs detected in the present study. Industrial POPs (e.g. PCBs and PBDEs) are in the low concentration range compared with other regions in the world. 6-OH-BDE-47 was the predominant congener of OH-PBDEs in both species, the toxicity effect make this compound call for concern.

In general, OCPs contamination is more severe in heron eggs whereas tern eggs were detected with higher levels of industrial chemicals. Hence, advanced environmental monitoring program should select proper water bird species in YRD in order to compare with other well-studied regions (e.g. PRD) and/or investigate bioaccumulation through food chain in aquatic ecosystem.

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