Environment International 130 (2019) 104952

Contents lists available at ScienceDirect

Environment International

journal homepage: www.elsevier.com/locate/envint

Spatial distribution and hazard of halogenated flame retardants and polychlorinated biphenyls to common kingfisher (*Alcedo atthis*) from a region of South China affected by electronic waste recycling

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ARTICLE INFO

Handling Editor: Yong-Guan Zhu

Keywords:
PBDEs
Alternative halogenated flame retardants
Electronic and electrical waste
Point-source effect
Waterbirds
Ecological hazard assessment

ABSTRACT

Numerous studies have reported bioaccumulation of halogenated flame retardants (HFRs) and polychlorinated biphenyls (PCBs) in wildlife from electronic waste (e-waste) recycling sites. However, the concentrations and hazards of HFRs and PCBs in wildlife from non–e-waste sites which were not involved in any known e-waste recycling activities in the e-waste–impacted region are still unclear. Polybrominated diphenyl ethers (PBDEs), alternative HFRs (AHFRs; including dechlorane plus, decabromodiphenyl ethane, and 1,2-bis(2,4,6-tribromophenoxy) ethane), and PCBs were quantified in common kingfishers (*Alcedo atthis*) from a region affected by e-waste recycling in South China, and potential adverse effects were evaluated. Concentrations of ΣPBDEs and ΣPCBs in kingfishers ranged from 2.1×10^3–1.3×10^5 ng/g lipid mass (lm) and 2.1×10^3–1.5×10^6 ng/g lm, respectively. At e-waste recycling sites, these concentrations were 100- to 1000-fold greater than those in kingfishers from non–e-waste areas, where concentrations of ΣPBDEs and ΣPCBs were 16–1.2×10^3 and 39–3.0×10^3 ng/g lm, respectively. Concentrations of ΣAHFRs in kingfishers from e-waste sites and non–e-waste sites ranged from 8.5 to 3.6×10^2 and 0.8–2.9×10^2 ng/g lm, respectively. The greatest concentrations of PCBs in kingfishers were measured from the e-waste sites. Additionally, kingfishers from four non–e-waste sites in the vicinity of e-waste sites had greater PCB concentrations compared to the other six non-e-waste sites. Concentrations of AHFRs were negatively and significantly correlated with distance from an e-waste site, which indicated that AHFRs from non–e-waste sites might be influenced by point sources. Further, a significant ($r^2 = 0.53$, $p = 0.02$) positive correlation between human population density and concentrations of ΣPBDEs in kingfishers from non–e-waste sites was observed. Concentrations of either PBDEs or PCBs from e-waste sites might pose severe, adverse reproductive effects to kingfishers, while the potential for adverse effects of PBDEs and PCBs to kingfishers from most non–e-waste sites seemed minimal.

1. Introduction

Generation of electronic and electrical waste (e-waste) has become a growing environmental concern as goods have become cheaper and product life-spans have decreased (McGrath et al., 2018). Approximately 30–50 million tons of e-waste are generated globally each year, with a growth rate of 3–5% per annum (Cucchiella et al., 2015). While currently regulated, previous dismantling of e-waste in some developing countries, such as China, Nigeria, and Ghana, resulted in significant releases of potentially toxic substances, including halogenated flame retardants (HFRs) and heavy metals (e.g., Cd, Cr, Hg, Zn and so on) (Wong et al., 2007; Leung et al., 2006) to which wildlife and humans can be exposed. HFRs, such as polybrominated diphenyl ethers (PBDEs), alternative halogenated flame retardants (AHFRs; including dechlorane plus (DP), decabromodiphenyl ethane (DBDPE), and 1,2-bis (2,4,6-tribromophenoxy) ethane (BTBPE)), as well as dielectrics such as polychlorinated biphenyls (PCBs), are generally persistent, bioaccumulative, and toxic, with the potential to cause adverse effects on both humans and wildlife (Darnerud, 2003). PBDEs and their metabolites have been shown to interfere with endocrine systems, including effects

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https://doi.org/10.1016/j.envint.2019.104952
Received 14 March 2019; Received in revised form 19 June 2019; Accepted 20 June 2019

to both reproductive hormone and thyroid hormone function (Jugan et al., 2010; Peng et al., 2016; Van den Steen et al., 2010). DP exposure might induce oxidative stress and alter detoxification and metabolic activities (Li et al., 2013; Wu et al., 2012). DBDPE and BTBPE have been shown to interfere with xenobiotic metabolism and thyroid hormone homeostasis in chicken embryos (Egloff et al., 2011). PCBs can cause mortality and adverse effects during development, such as deformities in early life stages (Doering et al., 2018; Giesy et al., 1994; Van den Berg et al., 1998).

China is the world's largest country for e-waste dumping, with importation of e-waste from developed countries reaching approximately 1.5 million tons in 2001 (Fu et al., 2018); the estimated e-waste importation is expected to be reduced to 0.32 million tons in 2018 (Zeng et al., 2016). Apart from transboundary importation, China itself is the second-largest global producer of e-waste and has been forecast to generate 15.5 and 28.4 million tons of e-waste in 2020 and 2030, respectively (Zeng et al., 2016). Guangdong Province is one of the most economically developed regions in China, which includes one of the largest manufacturing bases for electronic/electrical products (Pearl River Delta region, PRD) and two of the largest e-waste recycling sites in the world (Qingyuan and Guiyu) (Robinson, 2009). Over the past few decades, numerous studies have reported bioaccumulation of HFRs and PCBs in wildlife collected from the e-waste recycling sites in this region (Luo et al., 2009; Mo et al., 2013b; Wu et al., 2008), but the concentrations of HFRs and PCBs in wildlife from non-e-waste areas in the e-waste–impacted region were less clear.

Birds are generally widespread and sensitive to environmental changes, and they have been widely used to study environmental contamination and evaluate the health of certain ecosystems (Furness, 1993). PCBs can cause adverse effects on reproduction in exposed birds (Doering et al., 2018; Giesy et al., 1994; Van den Berg et al., 1998). The total abundance and species diversity of birds around e-waste recycling sites were reported to be inversely proportional to the severity of contamination by e-waste (Q. Zhang et al., 2014). However, limited information has been made available about the adverse effects and ecological hazards of HFRs and PCBs on wild birds in the wider e-waste–impacted region, e.g. liver histological changes of common kingfishers (*Alcedo atthis*) from nature reserves near a e-waste recycling site were found in this region (Wu et al., 2019). As regular resident, the common kingfisher have limited feeding ranges (10 km^2) and feed almost exclusively on small fish, it could be a suitable indictor for pollution of HFRs and PCBs.

In the present study, PBDEs, several AHFRs (including DBDPE, BTBPE, and DP), and PCBs were measured in the common kingfisher collected from two e-waste recycling sites and ten non–e-waste recycling sites in Guangdong Province, South China. Objectives of this study were to comprehensively investigate the magnitude and extent of concentrations of PBDEs, AHFRs, and PCBs in wild birds, represented by the kingfisher, in an e-waste recycling–impacted region. Spatial distributions of PBDEs, AHFRs, and PCBs in kingfishers were also investigated to examine the effects of point sources on surrounding areas. Finally, hazard quotients (HQs) were used to quantitatively evaluate the potential ecological risk of PBDEs and PCBs to kingfishers.

2. Materials and methods

2.1. Sample collection

A total of 138 adult kingfishers were collected from twelve sampling sites in Guangdong Province, South China between June 2012 and January 2015. The sampling sites included two e-waste recycling sites (e-waste sites) and ten sites were not involved in any known e-waste recycling activities (non–e-waste sites). Qingyuan (QY) and Guiyu (GY) are two known e-waste recycling sites in China. Ten non-e-waste sites, including Zengcheng (ZC), Zhaoqing (ZQ), Huizhou (HZ), Meizhou (MZ), Jiexi (JX), Heyuan (HY), Jiangmen (JM), Maoming (MM), Yingde (YD), and Shaoguan (SG) (Fig. S1 of Supporting information), were also considered. The biometric data of kingfishers include gender, body size, body weight and lipid content from 12 sampling sites were provided in Table S1 of Supporting information. Adult kingfishers were caught by plastic bird netting, as approved by the Forestry Bureau of Guangdong Province, China. In this study, birds were euthanized with N_2, the pectoral muscle from each bird was excised and stored at −20 °C until chemical analysis, and other tissues such as the liver and blood serum were specifically used for biochemical research in another study.

2.2. Sample extraction, cleanup, and chemical analysis

Methods for extraction, cleanup, identification, and quantification have been described in detail in a previous paper (Peng et al., 2015). Briefly, a homogenized sample of approximately 2–6 g breast muscle was mixed with ashed anhydrous sodium sulfate and spiked with surrogate standards (CBs 30, 65, and 204 for PCBs; $^{13}C_{12}$-BDE 209 and BDEs 77, 181, and 205 for PBDEs, DP, BTBPE, and DBDPE). Extraction was carried out with 180 mL of an acetone/hexane mixture (1:1, v/v) in a Soxhlet apparatus for 48 h. The extract was concentrated to 10 mL, and the lipid content was determined gravimetrically on an aliquot of 1 mL of extract. The remaining extract was concentrated to 2–3 mL, and then the lipids were removed by gel permeation chromatography (GPC). The GPC fraction containing target compounds was concentrated to 1–2 mL and further cleaned up on a multilayer column (10-mm inner diameter (i.d.)) packed with neutral activated silica (8 cm) and acidified silica gel (8 cm). Final extracts were concentrated to near dryness under a gentle stream of purified N_2 and reconstituted in 50 μL of isooctane. Internal standards (CBs 24, 82, and 198 for PCBs; BDEs 118 and 128 for PBDEs, DP, BTBPE, and DBDPE) were spiked prior to instrumental analyses. Target chemicals were analyzed by use of a gas chromatograph (GC) coupled with a mass spectrometer (MS). Detailed information on the instrumental conditions, quality assurance and control of chemical analysis are provided in the Supporting information.

2.3. Assessment of ecological hazards

A semi-probabilistic assessment was conducted in which hazards, as defined as the concentration affecting 5% of species (HC_5) or the 5th percentile of effect concentration (EC) values, were compared to point estimates of concentrations in tissues. HQs were used to quantitatively evaluate potential ecological risks to kingfishers from different sampling sites via exposure to PBDEs or PCBs (Su et al., 2014; R. Zhang et al., 2014). In this study, only hazards relating to effects on the reproduction of birds, including survival of embryos and piping success, eggshell thickness, and clutch size, were assessed. HQs are calculated by dividing the measured concentration (MEC) of contaminants in the muscle of kingfishers with the EC values, such as the predicted no effect concentration (PNEC) (Eq. (1)). Median concentrations of contaminants in the muscle of kingfishers from sampling sites were used as MECs.

$$\mathrm{HQ} = \frac{MECs}{PNECs} \times \text{Uncertainty Factor.} \quad (1)$$

For PBDEs, toxicological data related to the reproductive effects of PBDEs in individual levels of birds are scarce (Guigueno and Fernie, 2017) (Table S2), the 5th percentile of reported EC values to PBDEs were applied when estimating PNECs, using the lowest-observed EC (LOEC) instead of a no-observed EC (NOEC), so we used the uncertainty factor of 1 for $HQ_{05\ PBDEs}$ estimation.

For PCBs, the HC_5 values were used as PNEC values to estimate the risks posed by PCBs. Two HC_5 values were calculated based on two derived field-based species sensitivity distributions (SSDs) for birds relating breeding success exposure to PCBs from a previous study (Hoondert et al., 2018). One SSD derived from the field-based EC_{10} and another SSD derived from the field-based EC_{50} related to breeding

success were used to calculate the values of HQ_{10} and HQ_{50}, respectively. In considering the difference of tissue distribution and species sensitivity in birds, an uncertainty factor of 10 was assigned to encompass multiple potential variations. For interpretation, an HQ < 0.1 indicates no risk, 0.1–1 a low risk, 1–10 a moderate risk, and > 10 a high risk (modified from Lemly, 1996).

2.4. Data analyses

All concentrations were expressed on a lipid mass basis; concentrations with wet mass were provided Support information part for comparison purpose. For nondetects (below the method detection limits), half of the method detection limits were used to calculate the mean concentrations. ΣPBDEs, ΣDP, and ΣPCBs are defined as the sums of 14 BDE congeners, *syn*-DP and *anti*-DP, and 60 PCB congeners, respectively. ΣAHFRs refers to the sum of ΣDPs, DBDPE, and BTBPE. Concentration data in birds were tested for normality using the Kolmogorov–Smirnov one-sample test with Lilifor's transformation. Non-normally distributed data were logarithmically transformed to more closely approximate normal distributions. ANOVA and Tukey HSD post hoc tests were used to evaluate possible differences in concentrations among sampling sites. Distributions with statistically distinct profiles of HFRs and PCBs were grouped by unconstrained divisive cluster analyses (UncTree) of log-transformed data (Log (x + 1)) and statistical significance testing (SimProf; $\alpha = 0.05$) routines of Primer 7 (PRIMER-E Ltd., Ivybridge, U.K.). Euclidean-distance-resemblance matrices for detected contaminants were assessed by permutation-based, nonparametric, multivariate analyses (Primer 7). Other statistical analyses were performed with SPSS 18.0 software (SPSS Inc., Chicago, IL, USA) and Origin 9.0 (OriginLab Corporation, MA, USA) for Windows.

3. Results and discussion

3.1. Concentrations and profiles of congeners

3.1.1. PBDEs

PBDEs were detected in all samples, with the rates of detection of the individual congeners exceeding 87%. Concentrations of ΣPBDEs (the sum of 14 PBDE congeners) ranged from 2.1×10^2 to 1.3×10^5 and 16 to 1.2×10^3 ng/g lm in e-waste recycling sites and non–e-waste sites, respectively (Fig. 1, Tables S3, S4). Concentrations in kingfishers from locations with known rudimentary and intensive e-waste recycling activities were significantly greater (100- to 1000-fold) compared to those in the same region without known e-waste recycling activities ($F_{11,124} = 38.078$, $p < 0.001$; Tukey HSD: $p < 0.05$ for all cases). The greatest concentration of PBDEs was observed at GY, where previous studies have found abundant PBDEs in abiotic and biota samples (Leung et al., 2006; Li et al., 2018; Luo et al., 2009; Sun et al., 2012; Wu et al., 2008). These results indicated that intensive recycling of e-waste released large amounts of PBDEs into the environment. In non–e-waste sites, median concentrations of PBDEs in kingfishers ranged from 41 to 1.6×10^2 ng/g lm (Table S3) (mean: 47–3.1×10^2 ng/g lm), and kingfishers from ZC contained the greatest median concentration of 1.6×10^2 ng/g lm. Compared with reported concentrations in muscles of waterbirds from nonpoint pollution areas worldwide, PBDE concentrations in non–e-waste sites from this study were in the middle or higher end of the reported values (Table S5). For example, the geometric mean concentration of PBDE in guillemot (*Uria aalge*) from the Baltic Sea was 80 ng/g lm (Lundstedt-Enkel et al., 2006); the yellow-legged gull (*Larus michahellis*) and Euroasian coot (*Fulica atra*) from Isikli in Turkey had mean concentrations of 47 and 29 ng/g lm (Kocagoz et al., 2014), respectively. These results indicated that PBDEs are widespread in the environment in this study region.

BDE 47, BDE 100, BDE 153, BDE 154, BDE 202, and BDE 209 were predominant congeners in kingfishers, with mean contributions to total PBDE concentrations of 18%, 15%, 15%, 22%, 4.7%, and 9.4%, respectively (Fig. S2, Tables S6, S7), indicating that commercial formulations of penta-BDE, hexa-BDE, octa-BDE, and deca-BDE were their major sources. Greater concentrations of BDEs 47, 100, 153, and 154 might be attributed to bioaccumulation through the food chain of kingfishers (Mo et al., 2012). In addition, some evidence indicated that BDEs 100, 153, and 154 might have been derived from in vivo metabolism of BDE 209 by birds (Letcher et al., 2014).

3.1.2. AHFRs

Concentrations of ΣAHFRs in kingfishers from the e-waste sites and non–e-waste sites ranged from 8.5–3.6×10^2 ng/g lm and 0.8–2.9×10^2 ng/g lm, respectively (Table S3). Concentrations in kingfishers from e-waste sites were significantly greater compared to those at non–e-waste sites ($F_{11,119} = 28.494$, $p < 0.001$; Tukey HSD: $p < 0.05$ for all cases). ΣDP (the total concentration of *syn*-DP and *anti*-DP) was the most frequently detected AHFR, which was present in 100% of kingfishers, followed by DBDPE (94%), and BTBPE (71%).

Of the AHFRs assessed, DP was present at the greatest levels, with ΣDP concentrations ranging from 0.4 to 2.4×10^2 ng/g lm (Tables S3, S8, Fig. 1). ΣDP concentrations measured in kingfishers from e-waste sites (median concentrations in GY and QY were 1.7×10^2 and 57 ng/g lm, respectively) were significantly greater (10- to 1000-fold) than those in kingfishers from non–e-waste sites (median concentrations ranged from 0.9 to 4.8 ng/g lm, respectively; $F_{11,119} = 18.132$, $p < 0.001$; Tukey HSD: $p < 0.05$ for all cases). Greater concentrations of DP have been detected in air, dust, soils, plants, wildlife, and human blood serum and hair collected from e-waste sites (Mo et al., 2013b; Ren et al., 2009; Sverko et al., 2011; Wu et al., 2010; Zhang et al., 2011). Few studies have reported the occurrence of DP in waterbirds worldwide; most DP concentrations detected in wild birds have been reported in terrestrial birds and eggs. Compared to concentrations measured in the muscle of waterbirds in previous studies, concentrations of DP in kingfishers from e-waste sites were at the greater end of the reported range from polluted areas (Table S5). DP concentrations in kingfishers from non–e-waste sites in this study were comparable to previous values reported in kingfishers from a rural location in China (Mo et al., 2013b). These results indicated that e-waste recycling activities might accelerate the release of DP into local environments.

Concentrations of DBDPE and BTBPE in kingfishers ranged from 0.01 to 2.3×10^2 ng/g lm and non-detectable (nd) to 49 ng/g lm (Table S3, Fig. 1), respectively. Median concentrations of DBDPE in kingfishers from GY, ZC, and ZQ were 25, 18, and 23 ng/g lm, respectively, and they were significantly greater than those from the other locations in South China ($F_{11,117} = 17.380$, $p < 0.05$; Tukey HSD: $p < 0.05$ for all cases). The greatest concentration of DBDPE, 2.3×10^2 ng/g lm, was observed in an individual from ZC (Table S3). Compared to the median concentration in birds from hot spots, concentrations of DBDPE in kingfishers from GY, ZC, and ZQ were at the greater end of reported values (Table S3). E-waste activities in GY and industrial activities at PRD might contribute to the greater concentrations of DBDPE. Similar results were also observed for piscivorous birds from e-waste sites and in environments from industrialized areas (Chen et al., 2014; He et al., 2012; Luo et al., 2009). Furthermore, DBDPE was the most abundant AHFR in all non–e-waste sites except YD, with mean contributions to ΣAHFRs ranging from 52% to 89%. These results support the conclusion that DBDPE has become the major replacement of deca-BDE. Similar results have been observed in soils from other Asian countries (McGrath et al., 2018).

The median concentrations of BTBPE in kingfishers from two e-waste sites (GY and QY) were 9.0 and 0.6 ng/g lm. BTBPE levels in kingfishers from GY were significantly greater than in kingfishers from non–e-waste sites (nd to 0.2 ng/g lm) ($F_{11,117} = 2.383$, $p < 0.05$; Tukey HSD: $p < 0.05$ for all cases). BTBPE has been detected previously in waterbirds from e-waste sites, with median concentrations of 0.12, 3.3, and 1.9 ng/g lm in *G. cinerea*, *P. fusca*, and *G. striatus*, respectively (Table S5). Compared to these values, concentrations of

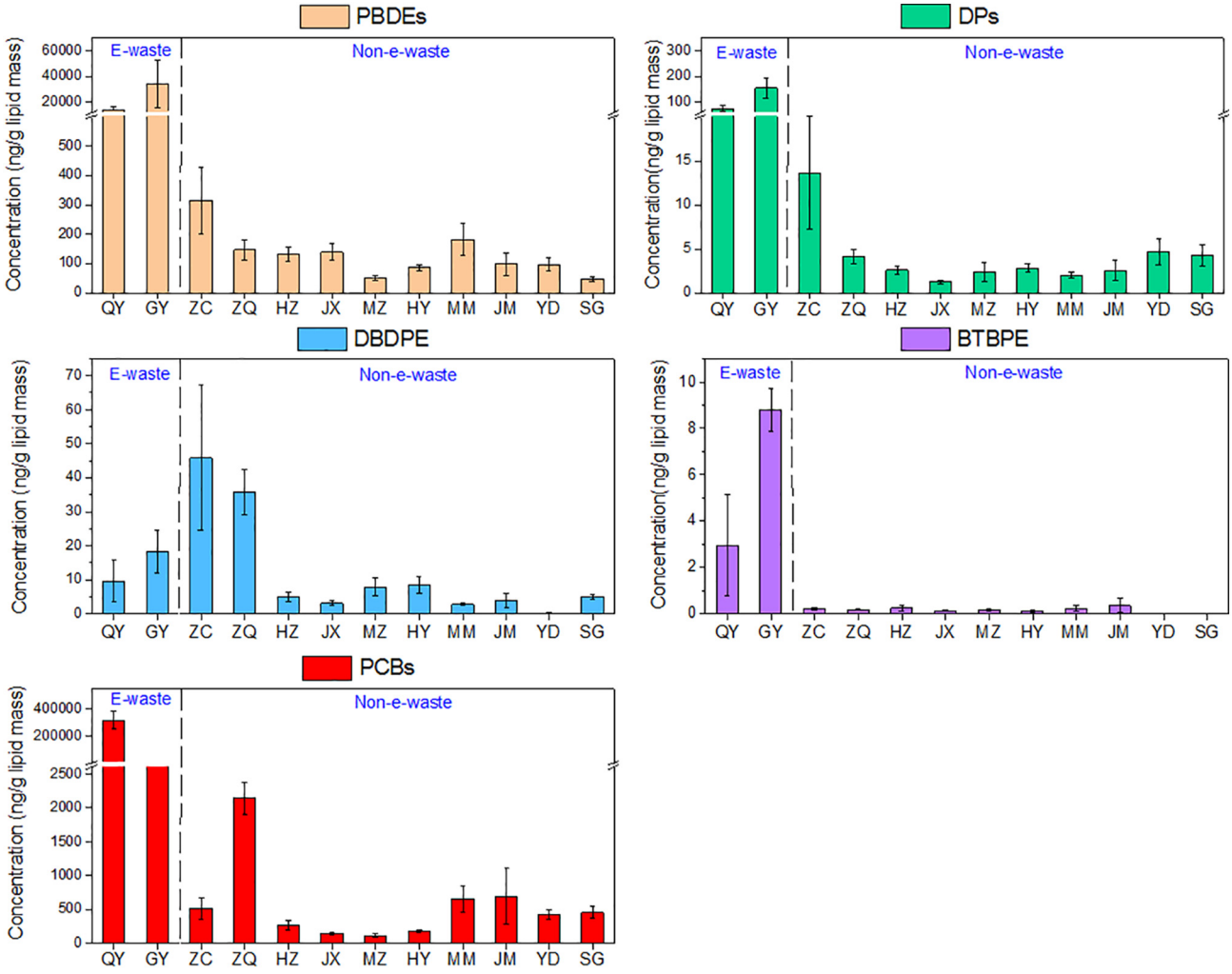

Fig. 1. Concentrations of HFRs (including PBDEs, DPs, DBDPEs, and BTBPEs) and PCBs in the muscle of kingfishers (*A. atthis*) collected from 12 locations in a region of South China affected by e-waste recycling. Error bar indicated the standard error (SE).

BTBPE in kingfishers from GY were among the greatest concentrations observed, which indicated that e-waste recycling activities are important sources of BTBPEs. BTBPE is utilized as a replacement for octa-BDE in a range of electrical parts, particularly in hard plastics used for outer casings like acrylonitrile butadiene styrene and high impact polystyrene from which BTBPE might be released during recycling of e-waste (McGrath et al., 2018).

A fraction of *anti*-DP (f_{anti}) was used to evaluate the possible stereoisomer selective enrichment of DP isomers, which was defined as the concentration of *anti*-DP divided by the total concentration of DP. In this study, median f_{anti} values of kingfishers from all sampling sites ranged from 0.61 to 0.88, while mean values ranged from 0.66 to 0.88 (Fig. S3), which were consistent with results of previous studies. Results of studies at various locations have demonstrated that f_{anti} values in biotic and abiotic matrices ranged from 0.64 to 0.80 (Hoh et al., 2006; Tomy et al., 2007; Wu et al., 2010). Except for one non–e-waste site (JM), the median f_{anti} value in kingfishers from e-waste recycling sites (QY and GY) was less than that of all other non–e-waste sites (Fig. S3). These results might be attributable to the dose-related accumulation of DP isomers in birds. Previous studies have reported a dose-dependent accumulation of DP isomers in the common quail (*Coturnix coturnix*), which exhibited lower f_{anti} values in individuals exposed to greater concentrations (Li et al., 2013).

3.1.3. PCBs

PCBs were detectable in all kingfishers from all sampling sites at concentrations from 39 to 1.5×10^6 ng/g lm for ΣPCBs (sum concentration of 60 PCB congeners) (Table S3). Median concentrations of ΣPCBs in kingfishers from e-waste recycling sites QY and GY were 2.6×10^5 and 9.7×10^3 ng/g lm (mean values: 3.2×10^5 and 1.1×10^4 ng/g lm, or 1.3×10^4 and 6.1×10^2 ng/g ww), respectively; which were 100- to 1000-fold greater than the concentrations in kingfishers from non–e-waste sites (Table S3) ($F_{11,119} = 8.636$, $p < 0.001$; Tukey HSD: $p < 0.05$ for all cases). Compared with the reported values in waterbirds from other contaminated countries (South Korea, Japan, Sweden, Turkey, and Romania), ΣPCB concentrations in this study are at the larger end (Table S5). In comparison, except ZQ, ΣPCB concentrations in the muscle tissue of kingfishers from non–e-waste sites (mean and median values ranged from 1.2×10^2 to 6.9×10^2 ng/g lm and 1.1×10^2 to 4.4×10^2 ng/g lm, respectively) were at the middle and upper end of reported ranges from nonpoint source areas worldwide (Table S5). Concentrations of PCBs in kingfishers from ZQ (median of 2.5×10^3 ng/g lm) were generally greater than other non–e-waste sites, which suggested potential PCB point pollution, such as dumping of abandoned electric capacitors from this site (Li, 2006). These results suggest that kingfishers from e-waste recycling sites and non–e-waste areas have been accumulating greater concentrations of PCBs, which might cause adverse health effects to the resident birds in this region.

The pattern of PCB congeners in kingfishers from various sampling sites are shown in Figs. S4 and S5. The predominant congeners in all kingfishers were CBs 153, 118, 138, and 180, with contributions of 25–39% to the total PCBs; this was consistent with the findings of previous studies (Jaspers et al., 2008; Mo et al., 2013a). These congeners are known to be slowly cleared from birds due to the presence of chlorine substitutions at meta-para positions on the phenyl rings (Drouillard et al., 2007). Tetra-, penta-, hexa-, and hepta-PCBs were the major homologue profiles at all sites and contributed > 82% to the total PCBs. Similar results have been reported in previous studies, where penta-, hexa-, and hepta-PCBs were the main homologues in birds (Dauwe et al., 2006; Yu et al., 2014). These homologue profiles might be related to the greater bioaccumulation potentials of those congeners. It has been suggested that HFRs and PCBs with values of log *K*ow 6–7 have a greater bioaccumulation ability, including penta-, hexa-, and hepta-PCBs (Kelly et al., 2007).

3.2. Spatial distribution and potential source effect

3.2.1. Spatial distribution

Profiles of PBDEs, AHFRs, and PCBs in kingfishers from 12 sampling sites were mainly clustered into four groups ($a = 0.05$) (Fig. 2). Group A included two e-waste recycling sites (QY and GY), which were clearly distinguished from all non–e-waste sites (Group B). Kingfishers from the e-waste area showed greater concentrations of PBDEs, DPs, and PCBs compared with non–e-waste sites (Fig. 1), which might be due to the primitive e-waste recycling and dismantling activities having released large amounts of contaminants into the ambient environment. Group B was clustered into three subgroups, including ZQ, ZC, and all other non–e-waste sites. PCBs were predominant contaminants in kingfishers from all non–e-waste sites with contributions of 59–92% to the total concentrations of contaminants, followed by PBDEs and AHFRs, with contributions of 6.7–46% and 0.7–6.2%, respectively (Fig. S6).

Kingfishers from ZQ accumulated greater concentrations of PCBs and DBDPEs, with CB28/31, CB66, CB99, CB101, CB118, CB138, and CB153/132 as the predominant congeners. Most of the congeners are tri-CBs, tetra-CBs, and penta-CBs, collectively contributing 76.7% to ΣPCBs. In China, tri-CBs (9000 tons) and penta- CBs (1000 tons) were produced from 1965 to 1974 (production of PCBs was banned in 1974) (China SEPA, 2003). Tri-, tetra-, and penta-CBs were the predominant compounds detected in soil from an electronic appliance disposal area in China (Zhou et al., 2018), consistent with the results of this study. A previous study reported that large quantities of PCBs have not been effectively disposed of after phasing out the use of PCBs in China (Li, 2006); this might related to the significant concentrations of PCB in ZQ. The greater concentrations of DBDPE (median: 23 ng/g lm) observed in kingfishers from ZQ are likely due to the fact that there are many industries in the PRD.

Starting in 2006, DBDPEs have been used as a replacement for FR-related products, such as deca-BDE in China with an annual production of 20,000 tons (Shi et al., 2009). Due to its larger molecular mass, DBDPE is less volatile and more bound to particles so that it does not migrate as far or as rapidly by atmospheric transport as some other chemicals, such as PCBs (Li et al., 2017). Thus, local FR-related industrial activities are likely sources that contribute to observed concentrations of DBDPE and deca-BDE in local wild birds.

Compared with other non–e-waste sites, greater concentrations of DBDPE and PBDEs were observed in kingfishers from ZC (Figs. 1, S2).

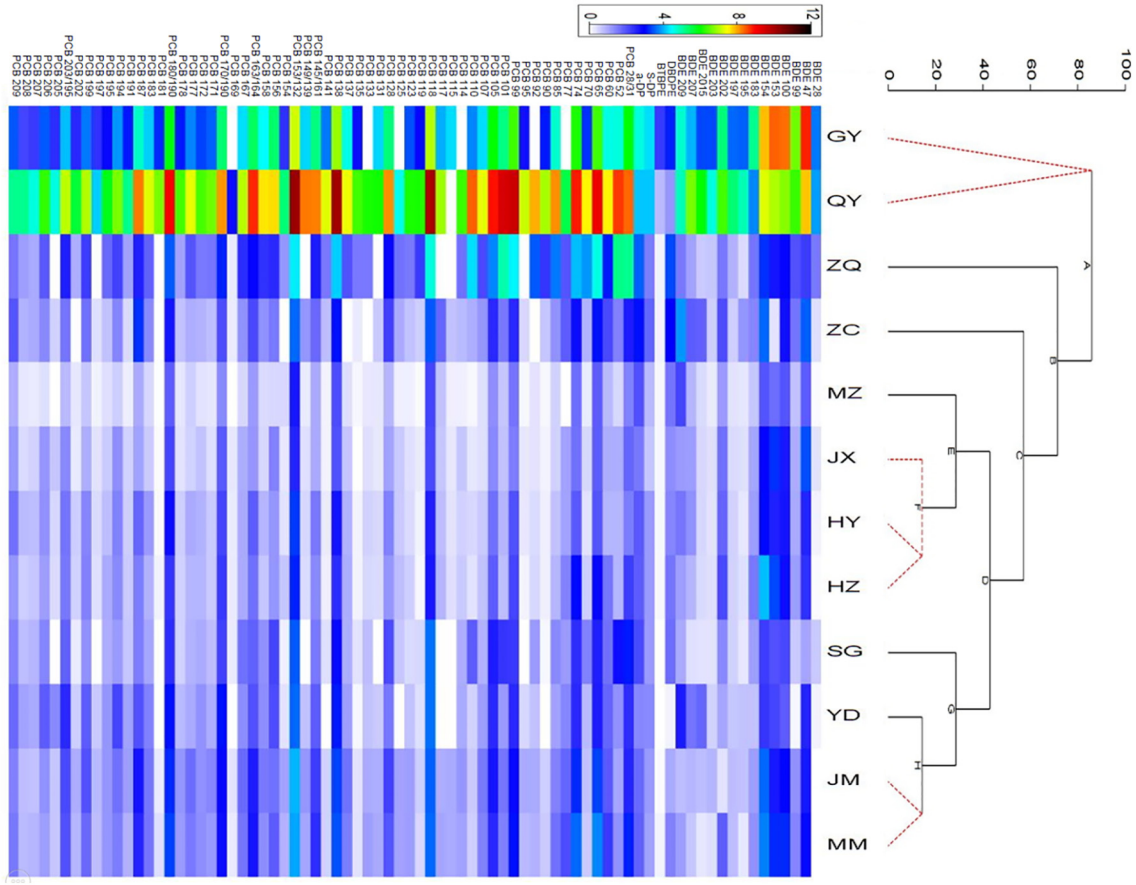

Fig. 2. Heatmap of concentrations (scale in ng/g lipid mass) of 78 contaminants in the muscle of kingfishers from 12 locations in a region of South China affected by e-waste recycling. Hierarchical cluster analysis was used with Ward's method and squared Euclidean distance. White indicates concentrations below method detection limits (MDLs).

BDE 209 was the major congener of PBDEs, with contributions of 30% to the total PBDEs. Similar results were observed for abiotic matrices, such as air, dust, and sediment from the PRD (Mai et al., 2005; Chen et al., 2014). ZC may be impacted by the industrial activities from industrialized urban areas, since it is located 50 km to the east of the capital of Guangdong Province, Guangzhou (GZ), where local industries in GZ can release DBDPE and PBDEs into the environment.

Except for ZQ and ZC, all other non–e-waste sites were clustered into two subgroups (Fig. S7). Group G from the northwest part of Guangdong Province, which included locations SG, YD, JM, and MM (Fig. S7), exhibited a relatively great abundance of PCBs compared with group E (75% to 88%) (Fig. S6). These four non–e-waste sites were closer to QY, where severe contamination with PCBs was observed in this study. Group E included MZ, JX, HY, and HZ from the southeast part of Guangdong Province and exhibited relatively large rates of detection of PBDEs compared with group G (29% to 46%) (Fig. S6). These four non–e-waste sites were closer to GY, where severe contamination with PBDEs was observed. These results indicate that the prevailing southeast and northeast winds in Guangdong Province might carry PCBs or PBDEs from the e-waste sites to these non–e-waste sites.

3.2.2. Potential source effect

For semi-volatile organic compounds (SVOCs), there is a common distribution pattern observed worldwide that is called "primary fractionation," which is a type of point-source fractionation (Li et al., 2017). To investigate potential sources of SVOCs, including PBDEs, some AHFRs, and PCBs, samples from non–e-waste sites were ranked by distance of the sample locations from the e-waste recycling site (QY or GY). Because greater concentrations of DBDPE in kingfishers were not only observed at e-waste sites but also non–e-waste sites (ZC and ZQ), ZC and ZQ were excluded. A log-linear relationship between concentrations of AHFRs in kingfishers and the distance from the source center (QY) was observed (Fig. 3), which demonstrated that concentrations of AHFRs in kingfishers decreased significantly as a function of distance from the source center. R values for regression models at e-waste recycling site QY for ΣAHFRs, BTBPEs, and DPs were −0.69, −0.66, and −0.88, respectively. These results suggest that e-waste recycling activities at QY have the potential to contaminate kingfishers in non–e-waste sites from this study region with AHFRs. Similar results were observed in previous studies, HFRs including DBDPE, BTBPE, and others decreased significantly in soils as a function of distance from the source centers (Li et al., 2017). Significant positive correlations between the natural logarithm of the concentration of PCBs in kingfishers and distance from the GY were observed (Fig. S8); it is speculated that the greater PCB concentrations in kingfishers from northeast portions of Guangdong Province, including locations ZC, YD, SG, JM, and MM, are influenced by the source center at QY. Legacy PCBs are influenced by "secondary fractionation," in which the transport of chemicals is mainly driven by temperature, soil organic carbon content, and, ultimately, surface–air partition coefficients (K_{SA}) for various environmental compartments or climate zones (Li et al., 2010). Prevailing southeast and northeast winds in Guangdong Province might influence the distribution of PCBs in this region with four non–e-waste sites in the vicinity of QY, which had greater concentrations of PCBs compared to other non–e-waste sites. This result contributes further evidence to support that processing of e-waste at QY was the dominant point-source of PCBs in the region.

Further, a significant (r^2 = 0.53, p = 0.02) positive correlation between concentrations of PBDEs in kingfishers and densities of populations of humans at 10 non–e-waste sites was observed (Fig. 4). This suggested that regions with greater populations of humans might have more use and disposal of flame retardant–containing products, thereby

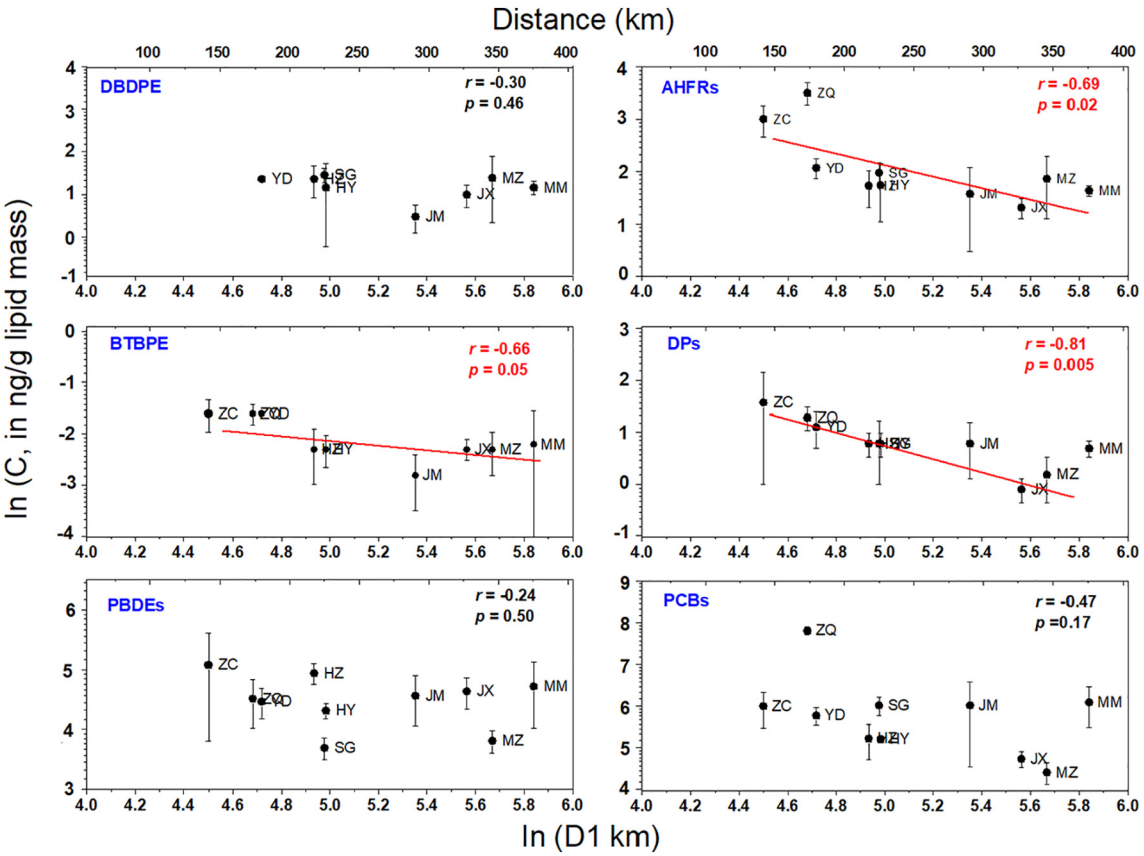

Fig. 3. Correlation analysis between the natural logarithm (ln) concentrations (ng/g lipid mass) of HFRs (including PBDEs, DPs, DBDPEs, and BTBPEs) and PCBs and the natural logarithm distance from e-waste recycling site QY (ln D1) (top x-axis: distance; bottom x-axis: logarithm distance).

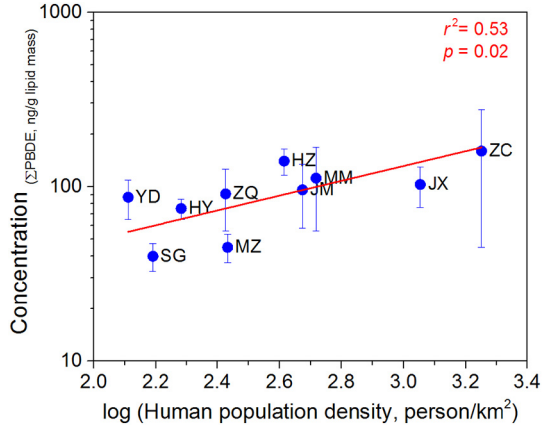

Fig. 4. Correlation analysis between concentrations of ΣPBDEs (median, ng/g lipid mass) and the logarithm human population density of ten non–e-waste sites.

releasing greater amounts of PBDEs into the environment and consequently leading to greater PBDE exposure in wildlife. This result was consistent with those of previous studies, in which concentrations of PBDEs in the eggs of gulls and starlings in Canada and passerine birds from South China were significantly and positively correlated to human population density (Chen et al., 2013; Sun et al., 2012).

For PBDEs, HQs from e-waste recycling sites and non–e-waste sites ranged from 5.4 to 2.2×10^2 and 2.2×10^{-2} to 2.7 (Fig. 5a, Table S9), respectively. Except for two non–e-waste sites, MZ and SG, mean HQ values for all other non–e-waste sites were > 0.1, indicating that PBDE contamination from most of the sampling sites may pose low-level hazards to the reproductive performance of birds.

HQ_{10} and HQ_{50} values of PCBs from e-waste recycling sites ranged from 0.4 to 1.5×10^2 and 2.8×10^{-2} to 10, respectively (Fig. 5b and Table S9). The mean HQ_{10} values for kingfishers from both e-waste sites, QY and GY, were > 1.0; meanwhile, the mean HQ_{50} values from the two e-waste sites were 2.3 and 0.2, respectively. The results may indicate that PCBs in this region might pose a risk to kingfisher reproduction. The relative ecological risk caused by PCBs was most likely due to point sources from the e-waste recycling activities, including storage of PCBs in environmental matrices that are re-emitted into the water column and subsequently accumulate in the food chain. HQ_{10} values and HQ_{50} values of PCBs from non–e-waste sites ranged from 0.6×10^{-2} to 0.2 and 0.4×10^{-3} to 1.5×10^{-2}, respectively. HQs for only one non–e-waste site, ZQ, were > 0.1. Thus, current concentrations of PCBs in kingfishers from non–e-waste areas are unlikely to cause reproductive impairment in this region. As dioxin-like compounds (DLCs), coplanar PCBs with greater potency of arylhydrocarbon receptors (AhRs) might cause adverse effects on the reproduction and development of birds and fish (Doering et al., 2018). To characterize the potential adverse effects to kingfishers exposed to coplanar PCBs, the toxic equivalent quantity (TEQ) concentrations of ten coplanar PCB congeners (including CBs 77, 81, 105, 114, 118, 123, 126, 156, 167, and 169) in kingfishers from 12 locations in South China were calculated based on the 2,3,7,8-tetrachlorodibenzo-*p*-dioxin (TCDD) equivalency factor for birds ($TEF_{WHO\text{-}avian}$) by the World Health Organization (Van den Berg et al., 1998). Calculated $TEQs_{PCBs}$ ranged from 0.1 to 4.9×10^3 pg/g wm (Fig. S9, Table S10). The greatest median TEQ concentration was observed in kingfishers from e-waste recycling sites (QY) with 7.2×10^2 pg/g wm. Compared to the toxicity reference values (TRVs) for embryo mortality and development impairment in birds exposed to TEQs, all TEQ_{PCBs} concentrations in kingfishers from QY and three kingfishers from non–e-waste sites exceeded the mean TRV (66 pg/g egg wm) in white leghorn chickens (*Gallus gallus*), which was the most sensitive species of bird (Suter, 2003). Despite PCBs in most non–e-waste sites being predicted to have no adverse effects on

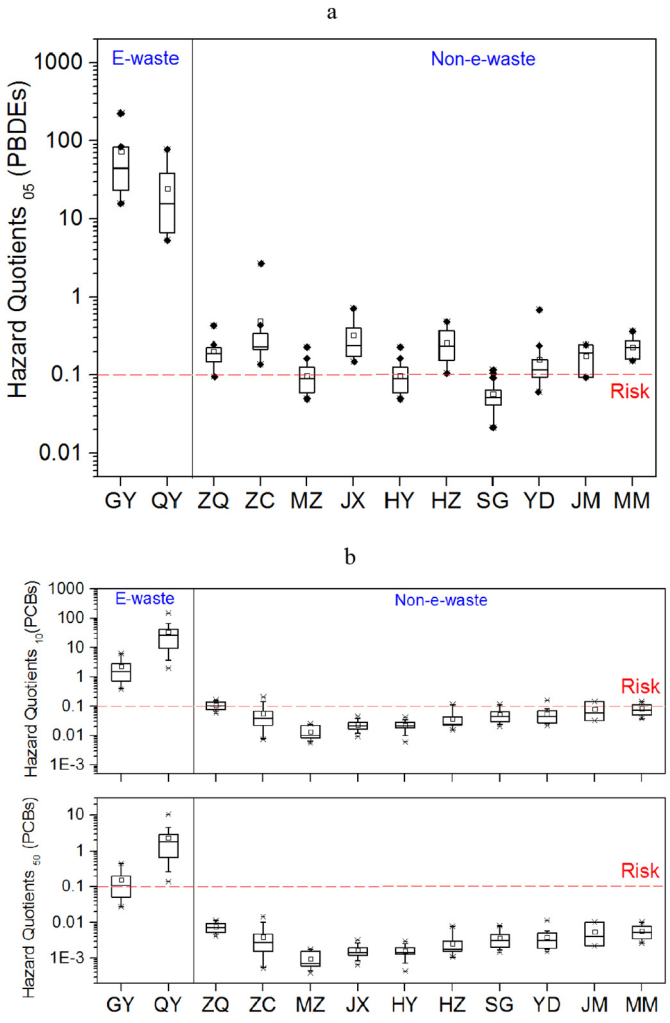

Fig. 5. Hazard quotients (HQs) in 12 sampling sites for exposure of kingfishers to PBDEs (a) and PCBs (b). Box plots are defined as follows: center line, median value; hallow square, mean value; box plot edges, the 25th and 75th percentiles.

kingfishers, exposure to multiple contaminants (PCBs, PBDEs, and AHFRs or other unknown compounds) might act additively or synergistically to affect the reproduction of wild birds. In particular, when the total potency of the effects modulated by AhR were considered, kingfishers from e-waste sites might be facing greater threats from DLCs regarding to the additively effects.

Several uncertainties limited the application of the resulting concentrations to a risk evaluation in the present study. For example, no toxicity threshold data on HFRs and PCBs are available for kingfishers, so EC estimates must be extrapolated from other bird species. To greatly reduce the probability of underestimating risk, the 5th percentile of EC values with PBDE and hazard concentrations of PCBs affecting 5% of species were calculated based on an SSD that was derived from field data on the reproductive success of more than eight bird species (Hoondert et al., 2018), and the resulting values were used for HQ evaluation. In addition, an uncertainty factor of 10 for tissue differences was applied to the risk estimate. In spite of the limitations, the preliminary reproductive hazards of legacy and emerging pollutants to wildlife in both e-waste sites and non–e-waste sites from the e-waste–impacted region were evaluated. Further studies should be addressed to the ecological hazards of PBDEs and PCBs on bird population levels at e-waste sites.

4. Conclusion

In this study, high levels of PBDEs, AHFRs, and PCBs in kingfishers from e-waste recycling sites were found. PBDEs are widespread in the environment in this study region. Kingfishers from non–e-waste sites with greater populations of humans have greater PBDE levels, since humans are increasingly releasing greater amounts of PBDEs in the environment. AHFRs from non–e-waste sites were negatively and significantly correlated with distance from an e-waste site. Further, kingfishers from non–e-waste sites in the vicinity of an e-waste site have greater PCB concentrations than do others. These results indicated that AHFRs and PCBs released from point sources, like the e-waste sites, have greater influence on the burden of these contaminants in kingfishers from surrounding areas. Existing PCB and PBDE concentrations at e-waste sites might pose severe adverse reproductive effects to kingfishers, while most non–e-waste sites are unlikely to cause adverse effects. Although previous dismantling of e-waste has been regulated, PBDEs and PCBs are still being released from known or unknown sources; additional monitoring will be necessary to evaluate the potential adverse effects of e-waste on kingfishers and other sensitive species.

Declaration of Competing Interest

None.

Acknowledgments

For support, we thank the National Natural Science Foundation of China [grant number 21707069], the Natural Science Foundation of Jiangsu Province [grant number BK20160650], National Basic Research Program of China [grant number 2015CB453102], Local Innovative and Research Teams Project of Guangdong Pearl River Talents Program [grant number 2017BT01Z134], and Key Research Program of Frontier Sciences, the Chinese Academy of Sciences [grant number QYZDJ-SSW-DQC018]. Prof. Giesy was supported by the "High Level Foreign Experts" program [grant number GDT20143200016] funded by the State Administration of Foreign Experts Affairs, the P.R. China to Nanjing University, and the Einstein Professor Program of the Chinese Academy of Sciences. He was also supported by the Canada Research Chair program and a Distinguished Visiting Professorship in the School of Biological Sciences of the University of Hong Kong. We also thank Ling Mo, Yanhong Zeng, Xiaobo Zheng, and Bin Tang from GIGCAS for assistance in the laboratory and field sampling.

Appendix A. Supplementary data

The supporting information includes detailed information regarding the method for target chemical analysis, quality assurance and control, figures (Figs. S1 to S9) and tables (Tables S1 to S10) showing locations of sampling sites, concentrations and compositional profiles of PBDEs, DPs, DBDPEs, and BTBPEs in kingfishers from the study area and other waterbirds worldwide, and the HQs and TEQs of contaminants in kingfishers. Supplementary data to this article can be found online at https://doi.org/10.1016/j.envint.2019.104952.

References

Chen, D., Martin, P., Burgess, N.M., Champoux, L., Elliott, J.E., Forsyth, D.J., Idrissi, A., Letcher, R.J., 2013. European Starlings (*Sturnus vulgaris*) suggest that landfills are an important source of bioaccumulative flame retardants to Canadian terrestrial ecosystems. Environ. Sci. Technol. 47, 12238–12247.

Chen, S.J., Ding, N., Zhu, Z.C., Tian, M., Luo, X.J., Mai, B.X., 2014. Sources of halogenated brominated retardants in house dust in an industrial city in southern China and associated human exposure. Environ. Res. 135C, 190–195.

SEPA, China, 2003. Building the capacity of the People's Republic of China to implement the Stockholm convention on POPs and develop a National implementation plan[R]. GEF Project Brief (GF/CPR/02/010), State Environmental Protection Administration (SEPA): China.

Cucchiella, F., D'Adamo, I., Koh, S.C.L., Rosa, P., 2015. Recycling of WEEEs: an economic assessment of present and future e-waste streams. Renew. Sust. Energ. Rev. 51, 263–272.

Darnerud, P.O., 2003. Toxic effects of brominated flame retardants in man and in wildlife. Environ. Int. 29, 841–853.

Dauwe, T., Jaspers, V.L.B., Covaci, A., Eens, M., 2006. Accumulation of organochlorines and brominated flame retardants in the eggs and nestlings of great tits, *Parus major*. Environ. Sci. Technol. 40, 5297–5303.

Doering, J.A., Wiseman, S., Giesy, J.P., Hecker, M., 2018. A cross-species quantitative adverse outcome pathway for activation of the aryl hydrocarbon receptor leading to early life stage mortality in birds and fishes. Environ. Sci. Technol. 52, 7524–7533.

Drouillard, K.G., Fernie, K.J., Letcher, R.J., Shutt, L.J., Whitehead, M., Gebink, W., Bird, D.A., 2007. Bioaccumulation and biotransformation of 61 polychlorinated biphenyl and four polybrominated diphenyl ether congeners in juvenile American kestrels (*Falco sparverius*). Environ. Toxicol. Chem. 26, 313–324.

Egloff, C., Crump, D., Chiu, S., Manning, G., McLaren, K.K., Cassone, C.G., Letcher, R.J., Gauthier, L.T., Kennedy, S.W., 2011. In vitro and in ovo effects of four brominated flame retardants on toxicity and hepatic mRNA expression in chicken embryos. Toxicol. Lett. 207, 25–33.

Fu, J., Zhang, H., Zhang, A., Jiang, G., 2018. E-waste recycling in China: a challenging field. Environ. Sci. Technol. 52, 6727–6728.

Furness, R., 1993. Birds as Monitors of Pollutants. Chapman and Hall, New York, NY, USA.

Giesy, J.P., Ludwig, J.P., Tillitt, D.E., 1994. Deformities in birds of the Great Lakes region. Assigning causality. Environ. Sci. Technol. 28, 128A–135A.

Guigueno, M.F., Fernie, K.J., 2017. Birds and flame retardants: a review of the toxic effects on birds of historical and novel flame retardants. Environ. Res. 154, 398–424.

He, M.J., Luo, X.J., Chen, M.Y., Sun, Y.X., Chen, S.J., Mai, B.X., 2012. Bioaccumulation of polybrominated diphenyl ethers and decabromodiphenyl ethane in fish from a river system in a highly industrialized area, South China. Sci. Total Environ. 419, 109–115.

Hoh, E., Zhu, L., Hites, R.A., 2006. Dechlorane plus, a chlorinated flame retardant, in the Great Lakes. Environ. Sci. Technol. 40, 1184–1189.

Hoondert, R.P.J., Hilbers, J.P., Hendriks, A.J., Huijbregts, M.A.J., 2018. Deriving field-based ecological risks for bird species. Environ. Sci. Technol. 52, 3716–3726.

Jaspers, V.L.B., Dirtu, A.C., Eens, M., Neels, H., Covaci, A., 2008. Predatory bird species show different patterns of hydroxylated polychlorinated biphenyls (HO-PCBs) and polychlorinated biphenyls (PCBs). Environ. Sci. Technol. 42, 3465–3471.

Jugan, M.L., Levi, Y., Blondeau, J.P., 2010. Endocrine disruptors and thyroid hormone physiology. Biochem. Pharmacol. 79, 939–947.

Kelly, B.C., Ikonomou, M.G., Blair, J.D., Morin, A.E., Gobas, F.A., 2007. Food web-specific biomagnification of persistent organic pollutants. Science 317, 236–239.

Kocagoz, R., Onmus, O., Onat, I., Cagdas, B., Siki, M., Orhan, H., 2014. Environmental and biological monitoring of persistent organic pollutants in waterbirds by non-invasive versus invasive sampling. Toxicol. Lett. 230, 208–217.

Lemly, A.D., 1996. Evaluation of the hazard quotient method for risk assessment of selenium. Ecotox. Environ. Safe. 35, 156–162.

Letcher, R.J., Marteinson, S.C., Fernie, K.J., 2014. Dietary exposure of American kestrels (*Falco sparverius*) to decabromodiphenyl ether (BDE-209) flame retardant: uptake, distribution, debromination and cytochrome P450 enzyme induction. Environ. Int. 63, 182–190.

Leung, A., Cai, Z.W., Wong, M.H., 2006. Environmental contamination from electronic waste recycling at Guiyu, southeast China. J Mater Cycles Waste 8, 13.

Li, L., 2006. Investigation and analysis of power equipments containing polychlorinated biphenyls in Guangdong. Guangdong Electric Power. 19, 2 (In Chinese).

Li, Y.F., Harner, T., Liu, L.Y., Zhang, Z., Ren, N.Q., Jia, H.L., Ma, J.M., Sverko, E., 2010. Polychlorinated biphenyls in global air and surface soil: distributions, air-soil exchange, and fractionation effect. Environ. Sci. Technol. 44, 2784–2790.

Li, Y., Yu, L.H., Zhu, Z.C., Dai, J.Y., Mai, B.X., Wu, J.P., Wang, J.S., 2013. Accumulation and effects of 90-day oral exposure to Dechlorane Plus in quail (*Coturnix coturnix*). Environ. Toxicol. Chem. 32, 1649–1654.

Li, W.L., Ma, W.L., Zhang, Z.F., Liu, L.Y., Song, W.W., Jia, H.L., Ding, Y.S., Nakata, H., Minh, N.H., Sinha, R.K., Moon, H.B., Kannan, K., Sverko, E., Li, Y.F., 2017. Occurrence and source effect of novel brominated flame retardants (NBFRs) in soils from five Asian countries and their relationship with PBDEs. Environ. Sci. Technol. 51, 11126–11135.

Li, N., Chen, X.-W., Deng, W.-J., Giesy, J.P., Zheng, H.-L., 2018. PBDEs and dechlorane plus in the environment of Guiyu, Southeast China: a historical location for e-waste recycling (2004, 2014). Chemosphere 199, 603–611.

Lundstedt-Enkel, K., Asplund, L., Nylund, K., Bignert, A., Tysklind, M., Olsson, M., Orberg, J., 2006. Multivariate data analysis of organochlorines and brominated flame retardants in Baltic Sea guillemot (*Uria aalge*) egg and muscle. Chemosphere 65, 1591–1599.

Luo, X.J., Zhang, X.L., Liu, J., Wu, J.P., Luo, Y., Chen, S.J., Mai, B.X., Yang, Z.Y., 2009. Persistent halogenated compounds in waterbirds from an e-waste recycling region in South China. Environ. Sci. Technol. 43, 306–311.

Mai, B.X., Chen, S.J., Luo, X.J., Chen, L.G., Yang, Q.S., Sheng, G.Y., Peng, P.G., Fu, J.M., Zeng, E.Y., 2005. Distribution of polybrominated diphenyl ethers in sediments of the Pearl River Delta and adjacent South China sea. Environ. Sci. Technol. 39, 3521–3527.

McGrath, T.J., Morrison, P.D., Ball, A.S., Clarke, B.O., 2018. Spatial distribution of novel and legacy brominated flame retardants in soils surrounding two Australian electronic waste recycling facilities. Environ. Sci. Technol. 52, 474–483.

Mo, L., Wu, J.P., Luo, X.J., Zou, F.S., Mai, B.X., 2012. Bioaccumulation of polybrominated diphenyl ethers, decabromodiphenyl ethane, and 1,2-bis(2,4,6-tribromophenoxy) ethane flame retardants in kingfishers (*Alcedo atthis*) from an electronic waste-

recycling site in South China. Environ. Toxicol. Chem. 31, 2153–2158.
Mo, L., Wu, J.P., Luo, X.J., Li, K.L., Peng, Y., Feng, A.H., Zhang, Q., Zou, F.S., Mai, B.X., 2013a. Using the kingfisher (*Alcedo atthis*) as a bioindicator of PCBs and PBDEs in the dinghushan biosphere reserve, China. Environ. Toxicol. Chem. 32, 1655–1662.
Mo, L., Wu, J.P., Luo, X.J., Sun, Y.X., Zheng, X.B., Zhang, Q., Zou, F.S., Mai, B.X., 2013b. Dechlorane Plus flame retardant in kingfishers (*Alcedo atthis*) from an electronic waste recycling site and a reference site, South China: influence of residue levels on the isomeric composition. Environ. Pollut. 174, 57–62.
Peng, Y., Wu, J.P., Tao, L., Mo, L., Tang, B., Zhang, Q., Luo, X.J., Zou, F.S., Mai, B.X., 2015. Contaminants of legacy and emerging concern in terrestrial passerines from a nature reserve in South China: residue levels and inter-species differences in the accumulation. Environ. Pollut. 203, 7–14.
Peng, Y., Xia, P., Zhang, J.J., Villeneuve, D.L., Zhang, J.M., Wang, Z.H., Wei, S., Yu, H.X., Zhang, X.W., 2016. Toxicogenomic assessment of 6-OH-BDE47 induced developmental toxicity in chicken embryos. Environ. Sci. Technol. 50, 12493–12503.
Ren, G.F., Yu, Z.Q., Ma, S.T., Li, H.R., Peng, P.G., Sheng, G.Y., Fu, J.M., 2009. Determination of dechlorane plus in serum from electronics dismantling workers in South China. Environ. Sci. Technol. 43, 9453–9457.
Robinson, B.H., 2009. E-waste: an assessment of global production and environmental impacts. Sci. Total Environ. 408, 183–191.
Shi, T., Chen, S.J., Luo, X.J., Zhang, X.L., Tang, C.M., Luo, Y., Ma, Y.J., Wu, J.P., Peng, X.Z., Mai, B.X., 2009. Occurrence of brominated flame retardants other than polybrominated diphenyl ethers in environmental and biota samples from southern China. Chemosphere 74, 910–916.
Su, H., Wu, F., Zhang, R., Zhao, X., Mu, Y., Feng, C., Giesy, J.P., 2014. Toxicity reference values for protecting aquatic birds in China from the effects of polychlorinated biphenyls. In: Whitacre, D.M. (Ed.), Reviews of Environmental Contamination and Toxicology Volume: With Cumulative and Comprehensive Index Subjects Covered. Volumes 221–230. Springer International Publishing, Cham, pp. 59–82.
Sun, Y.X., Luo, X.J., Mo, L., Zhang, Q., Wu, J.P., Chen, S.J., Zou, F.S., Mai, B.X., 2012. Brominated flame retardants in three terrestrial passerine birds from South China: geographical pattern and implication for potential sources. Environ. Pollut. 162, 381–388.
Suter, G.W., 2003. Analysis of laboratory and field studies of reproductive toxicity in birds exposed to dioxin-like compounds for use in ecological risk assessment. Vol. 114. EPA/600/R-03.
Sverko, E., Tomy, G.T., Reiner, E.J., Li, Y.F., McCarry, B.E., Arnot, J.A., Law, R.J., Hites, R.A., 2011. Dechlorane plus and related compounds in the environment: a review. Environ. Sci. Technol. 45, 5088–5098.
Tomy, G.T., Pleskach, K., Ismail, N., Whittle, D.M., Helm, P.A., Sverko, E., Zaruk, D., Marvin, C.H., 2007. Isomers of dechlorane plus in Lake Winnipeg and Lake Ontario food webs. Environ Sci Technol 41, 2249–2254.
Van den Berg, M., Birnbaum, L., Bosveld, A.T.C., Brunstrom, B., Cook, P., Feeley, M., Giesy, J.P., Hanberg, A., Hasegawa, R., Kennedy, S.W., Kubiak, T., Larsen, J.C., van Leeuwen, F.X.R., Liem, A.K.D., Nolt, C., Peterson, R.E., Poellinger, L., Safe, S., Schrenk, D., Tillitt, D., Tysklind, M., Younes, M., Waern, F., Zacharewski, T., 1998. Toxic equivalency factors (TEFs) for PCBs, PCDDs, PCDFs for humans and wildlife. Environ. Health Perspect. 106, 775–792.
Van den Steen, E., Eens, M., Geens, A., Covaci, A., Darras, V.M., Pinxten, R., 2010. Endocrine disrupting, haematological and biochemical effects of polybrominated diphenyl ethers in a terrestrial songbird, the European starling (*Sturnus vulgaris*). Sci. Total Environ. 408, 6142–6147.
Wong, M.H., Wu, S.C., Deng, W.J., Yu, X.Z., Luo, Q., Leung, A.O.W., Wong, C.S.C., Luksemburg, W.J., Wong, A.S., 2007. Export of toxic chemicals - a review of the case of uncontrolled electronic-waste recycling. Environ. Pollut. 149, 131–140.
Wu, J.P., Luo, X.J., Zhang, Y., Luo, Y., Chen, S.J., Mai, B.X., Yang, Z.Y., 2008. Bioaccumulation of polybrominated diphenyl ethers (PBDEs) and polychlorinated biphenyls (PCBs) in wild aquatic species from an electronic waste (e-waste) recycling site in South China. Environ. Int. 34, 1109–1113.
Wu, J.P., Zhang, Y., Luo, X.J., Wang, J., Chen, S.J., Guan, Y.T., Mai, B.X., 2010. Isomer-specific bioaccumulation and trophic transfer of Dechlorane Plus in the freshwater food web from a highly contaminated site, South China. Environ. Sci. Technol. 44, 606–611.
Wu, B., Liu, S., Guo, X.C., Zhang, Y., Zhang, X.X., Li, M., Cheng, S.P., 2012. Responses of mouse liver to dechlorane plus exposure by integrative transcriptomic and metabonomic studies. Environ. Sci. Technol. 46, 10758–10764.
Wu, J.P., Peng, Y., Zhi, H., Chen, X.Y., Wu, S.K., Tao, L., Zeng, Y.H., Luo, X.J., Mai, B.X., 2019. Contamination of organohalogen chemicals and hepatic steatosis in common kingfisher (*Alcedo atthis*) breeding at a nature reserve near e-waste recycling sites in South China. Sci. Total Environ. 659, 561–567.
Yu, L.H., Luo, X.J., Liu, H.Y., Zeng, Y.H., Zheng, X.B., Wu, J.P., Yu, Y.J., Mai, B.X., 2014. Organohalogen contamination in passerine birds from three metropolises in China: geographical variation and its implication for anthropogenic effects on urban environments. Environ. Pollut. 188, 118–123.
Zeng, X., Gong, R., Chen, W.Q., Li, J., 2016. Uncovering the recycling potential of "New" WEEE in China. Environ. Sci. Technol. 50, 1347–1358.
Zhang, X.L., Luo, X.J., Liu, H.Y., Yu, L.H., Chen, S.J., Mai, B.X., 2011. Bioaccumulation of several brominated flame retardants and Dechlorane Plus in waterbirds from an e-waste recycling region in South China: associated with trophic level and diet sources. Environ. Sci. Technol. 45, 400–405.
Zhang, Q., Wu, J., Sun, Y., Zhang, M., Mai, B., Mo, L., Lee, T.M., Zou, F., 2014. Do bird assemblages predict susceptibility by e-waste pollution? A comparative study based on species-and guild-dependent responses in China agroecosystems. PLoS One 10, e0122264.
Zhang, R., Guo, J., Wu, F., Mu, Y., Giesy, J.P., Chang, H., Zhao, X., Feng, C., 2014. Toxicity reference values for polybrominated diphenyl ethers: risk assessment for predatory birds and mammals from two Chinese lakes. Rev. Environ. Contam. Toxicol. 229, 111–137.
Zhou, Y.H., Yin, G., Du, X.Y., Xu, M.Y., Qiu, Y.L., Ahlqvist, P., Chen, Q.F., Zhao, J.F., 2018. Short-chain chlorinated paraffins (SCCPs) in a freshwater food web from Dianshan Lake: occurrence level, congener pattern and trophic transfer. Sci. Total Environ. 615, 1010–1018.

Supporting Information

Spatial distributions and hazard of halogenated flame retardants and polychlorinated biphenyls to common kingfisher (Alcedo atthis) from a region of South China affected by electronic waste recycling

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Supplementary Methods

Target chemical analysis

PCBs were analyzed by use of an Agilent 6890 gas chromatograph (GC) coupled with an Agilent series 5975B mass spectrometer (MS). The MS was operated in electron impact and selected ion monitoring (SIM) modes. A DB-5 MS capillary column (60 m length, 250-μm i.d., 0.25-μm film thickness; J&W Scientific) was used for separation of individual PCB congeners. Tri- to hepta-BDE congeners (BDEs 28, 47, 66, 85, 100, 99, 153, 154, and 183) and DP isomers were analyzed using an Agilent 6890 GC–5975 MS operated in electron capture negative ionization (ECNI) mode. A DB-XLB capillary column (30-m length, 250-μm i.d., 0.25-μm film thickness; J&W Scientific) was used for separation of congeners. Octa- to deca-BDE congeners (BDEs 196, 197, 202, 203, 206, 207, 208, and 209), DBDPE, and BTBPE were performed using a Shimadzu Model QP2010 GC–MS by use of ECNI in SIM mode. A DB-5HT capillary column (15-m length, 250-μm i.d., 0.1-μm film thickness; J&W Scientific) was used for separation. Details of GC conditions and monitored ions have been described previously (Mo et al., 2012; Peng et al., 2015).

Quality assurance and control

Procedural blanks, spiking blanks, and blind triplicate samples were processed for quality assurance. A procedural blank was processed in each batch (n = 11) of samples. Traces of BDEs 47, 206, 207, and 209, *anti*-DP, and CBs 28/31, 118, 128, 153, and 180/193 were detected in procedural blanks (0.9–8.5, 0.5–0.7, and 0.2–2.0 ppb for PBDEs, DPs, and PCBs, respectively), and the final reported concentrations were corrected for blank backgrounds. Recoveries of target compounds were evaluated by spiking known concentrations of BDEs 77, 181, and 205, ^{13}C-BDE 209,

and CBs 30, 65, and 204 as surrogates in the samples and by spiking known amounts of 20 major PCB congeners and 10 major PBDE congeners in solution. All surrogates passed through the entire analytical procedure. Mean recoveries of surrogate standards in samples (mean ± standard error) were 97% ± 10%, 92% ± 8.2%, 87% ± 17% and 80% ± 16% for BDE 77, 181, 205, and ^{13}C-BDE 209, and 85% ± 17%, 94% ±12%, and 92% ± 13% for CB 30, 65, and 204, respectively. Mean recoveries of spiked PBDEs, DBDPE and BTBPE, DPs, and PCBs in solution ranged from 83% to 105%, 90% to 107%, 88% to 104%, and 80% to 118%, respectively. Reported concentrations were not corrected for recoveries of surrogates. Triplicate samples of muscle were analyzed, and relative standard deviations of target compounds in triplicate samples were less than 13%. Instrumental quality control was performed by regular injection of solvent blanks and standard solutions. On-column degradations of BDE209 were checked by daily injection of BDE 209 standard solutions before sample analysis. Degradation of BDE 209 on the GC liner was observed, but degradation was < 3%.

Method detection limits (MDLs) were estimated as means of the target compounds detected in procedure blanks plus three times their standard deviations. For compounds undetected in procedural blanks, MDLs were defined as a signal-to-noise ratio (S/N ratio) of 10. Based on a mean of 0.04 g lipid in the samples, MDLs for PCBs and PBDEs ranged from 0.1 to 1.2 and 0.1 to 1.0 ng/g lipid mass (lm), respectively. MDLs for DBDPE and BTBPE were 1.2 and 0.01 ng/g lm, respectively. MDLs for *syn*-DP and *anti*-DP were 0.1 and 0.9 ng/g lm, respectively.

Table S1. Biometric data of the common kingfisher collected from 12 locations in a region of South China affected by e-waste recycling.

Locations	N	Female:Male	Body length (cm)	Body weight (g)	Lipid content (%)
E-waste sites					
QY	24	12:12	17.1 (16.5–17.6) [a]	22.8 (19.5–28.6)	4.2 (2.6–6.9)
GY	6	4:2	17.2 (16.9–17.8)	25.3 (23.4–28.7)	5.6 (4.0–6.9)
Non–e-waste sites					
ZC	10	2:8	16.1 (15.3–16.9)	21.7 (16.6–27.0)	4.0 (1.1–8.3)
ZQ	10	2:11	17.0 (15.0–18.0)	24.0 (18.0–28.0)	3.2 (2.3–4.5)
HZ	9	3:6	16.8 (16.5–18.0)	23.7 (20.3–25.5)	4.7 (3.5–5.4)
JX	12	7:5	16.5 (15.8–17.2)	24.5 (21.4–27.1)	5.4 (3.8–6.8)
MZ	15	6:9	16.9 (16.1–18.0)	23.2 (19.9–28.7)	5.0 (3.5–8.3)
HY	13	6:7	17.4 (16.5–18.1)	24.2 (21.6–27.1)	4.9 (3.4–6.8)
JM	3	1:2	16.3 (15.2–17.0)	22.1 (19.0–25.1)	4.7 (3.3–6.3)
MM	8	5:3	17.0 (15.7–17.7)	22.1 (20.2–23.5)	5.2 (3.5–7.5)
YD	15	2:13	16.7 (12.6–18.0)	22.5 (20.6–25.0)	3.9 (2.8–5.8)
SG	13	2:13	17.0 (12.0–18.0)	21.0 (19.0–26.0)	3.0 (2.1–4.5)

[a] Mean values (range).

Table S2. Thresholds of effects of PBDEs on the reproduction of wild birds.

Chemicals	Threshold levels (lowest-observed-effect-level, LOEL)	Species	Effect related reproductive success	References
PBDEs	1.8×10^3 ng/g egg wm [a] or 3.2×10^3 ng/g egg lm [b]	American kestrel (*Falco sparverius*)	impaired pipping and hatching success	McKernan et al. 2009
Penta-BDE Mixture，DE 71	2.9×10^2 ng/g egg wm	American kestrel (*Falco sparverius*)	Eggshell thinning	Fernie et al., 2009
PBDEs	1.5×10^2 mg sum PBDEs/bird (85 g per bird)= 1.8×10^3 ug/g wm	European starlings (*Sturnus vulgaris*)	reproductive performance	Van den Steen et al., 2009
BDE-17, -28, -47, -49, -66, -85, -99, -100, -138, -153, -154, -183, -190, -209.	1.0×10^3 ng/g egg wm	Osprey (*Falco sparverius*)	Negative relationship between productivity and ∑PBDE concentration	Henny et al., 2009
BDE-47, -99, -100, -153, -154.	5.9×10^2or 2.0×10^2 ng/g wm	Tree swallows (*Tachycineta bicolor*)	influence the timing of egg laying	Gilchrist et al., 2014

[a] wet mass.

[b] lipid mass.

Table S3. Medians and ranges of concentrations (ng/g lipid mass) of PBDEs, AHFRs (including DPs, DBDPEs, and BTBPEs), and PCBs in the muscle of kingfishers from 12 locations in a region of South China affected by e-waste recycling.

Locations	N	Lipid (%) [a]	ΣPBDEs [b]	AHFRs			ΣPCBs [d]
				ΣDP [c]	DBDPE	BTBPE	
E-waste sites							
QY	24	4.2 (2.6–6.9)	9.7×10^3 (2.1×10^3–4.9×10^4)	57 (13–2.4×12^3)	2.4 (0.3–1.1×10^2)	0.6 (nd [e]–49)	2.6×10^5 (2.1×10^4–1.5×10^6)
GY	6	5.6 (4.0–6.9)	1.9×10^4 (6.9×10^3–1.3×10^5)	1.7×10^2 (19–2.2×10^2)	25 (20–39)	9.0 (5.6–12)	9.7×10^3 (2.1×10^3–2.5×10^4)
Non–e-waste sites							
ZC	10	4.0 (1.1–8.3)	1.6×10^2 (59–1.2×10^3)	4.8 (1.2–65)	18 (4–2.3×10^2)	0.2 (nd–0.5)	4.0×10^2 (60–1.5×10^3)
ZQ	10	3.2 (2.3–4.5)	90 (50–4.1×10^2)	3.9 (1.0–13)	23 (0.4–90)	0.2 (0.04–0.4)	2.5×10^3 (7.1×10^2–3.0×10^3)
HZ	15	5.0 (3.5–8.3)	1.4×10^2 (46–2.4×10^2)	2.2 (0.9–5.7)	3.9 (1.1–15)	0.1 (0.02–1.3)	1.8×10^2 (1.1×10^2–7.9×10^2)
JX	12	5.4 (3.8–6.8)	1.0×10^2 (64–3.1×10^2)	0.9 (0.4–2.5)	2.7 (0.01–8.7)	0.1 (nd–0.2)	1.1×10^2 (72–2.9×10^2)
MZ	13	4.9 (3.4–6.8)	45 (16–1.4×10^2)	1.2 (0.4–17)	4.0 (1.6–34)	0.1 (0.02–0.7)	1.1×10^2 (72–2.9×10^2)
HY	9	4.7 (3.5–5.4)	75 (23–1.4×10^2)	2.2 (0.7–6.5)	3.2 (2.3–13)	0.1 (nd–0.4)	1.8×10^2 (39–2.5×10^2)
JM	13	3.0 (2.1–4.5)	96 (34–1.7×10^2)	2.2 (0.7–4.7)	1.6 (1.6–8.2)	0.06 (nd–0.9)	4.1×10^2 (1.6×10^2–1.5×10^3)
MM	15	3.9 (2.8–5.8)	1.1×10^2 (32–4.2×10^2)	2.0 (0.7–3.6)	3.2 (0.8–4.4)	0.2 (nd–1.0)	4.4×10^2 (2.5×10^2–1.9×10^3)
YD	3	4.7 (3.3–6.3)	70 (25–1.9×10^2)	2.8 (0.7–24)	3.9 (1.2–24)	0.2 (nd–1.7)	3.1×10^2 (1.5×10^2–1.0×10^3)
SG	8	5.2 (3.5–7.5)	41 (19–92)	2.2 (0.8–12)	4.3 (1.9–9.7)	nd	4.1×10^2 (1.8×10^2–1.3×10^3)

[a] Mean (range).

[b] Sum of 14 BDE congeners.

[c] Sum of *syn*-DP and *anti*-DP concentrations.

[d] Sum of 60 CB congeners.

[e] Not detectable.

Table S4. Medians and ranges of concentrations (ng/g wet mass) of PBDEs, AHFRs (including DPs, DBDPEs, and BTBPEs), and PCBs in the muscle of kingfishers from 12 locations in a region of South China affected by e-waste recycling.

Locations	ΣPBDEs [a]	AHFRs			ΣPCBs [c]
		ΣDP [b]	DBDPE	BTBPE	
E-waste sites					
QY	3.5×10^{2} (1.2×10^{2}–1.7×10^{3})	2.6 (0.6–6.8)	8.9×10^{-2} (0.3–2.3×10^{2})	5.9×10^{-2} (nd [d]–1.9)	1.1×10^{4} (7.4×10^{2}–5.9×10^{4})
GY	1.0×10^{3} (3.5×10^{2}–5.1×10^{3})	9.1 (1.3–15)	1.4 (0.3–1.8)	0.4 (0.4–0.8)	4.3×10^{2} (1.5×10^{2}–1.7×10^{3})
Non–e-waste sites					
ZC	5.2 (3.1–60)	0.2 (4.8×10^{-2}–1.9)	0.6 (0.3–6.7)	6.1×10^{-3} (nd–1.0×10^{-2})	14 (3.2–46)
ZQ	3.0 (2.2–9.0)	0.1 (1.3×10^{-2}–0.2)	1.0 (0.5–2.7)	0.5×10^{-2} (0.1×10^{-2}–1.0×10^{-2})	80 (19–1.4×10^{2})
HZ	5.4 (2.4–11)	2.2 (0.9–5.7)	0.2 (5.0×10^{-2}–0.8)	4.9×10^{-2} (0.8×10^{-3}–4.5×10^{-2})	8.4 (5.0–41)
JX	5.5 (3.4–16)	4.9×10^{-2} (2.6×10^{-2}–0.2)	0.1 (6.7×10^{-4}–4.7)	5.4×10^{-3} (nd–1.2×10^{-2})	6.6 (3.4–16)
MZ	2.0 (1.1–5.2)	5.8×10^{-2} (3.0×10^{-2}–0.6)	0.2 (0.1–1.6)	4.2×10^{-3} (2.0×10^{-3}–2.4×10^{-2})	3.7 (2.7–12)
HY	3.5 (1.6–8.2)	0.1 (4.4×10^{-2}–0.3)	0.2 (8.9×10^{-2}–1.0)	3.6×10^{-3} (nd–1.5×10^{-2})	7.6 (2.6–16)
JM	4.4 (2.1–5.5)	0.1 (4.6×10^{-2}–0.2)	0.1 (7.4×10^{-2}–0.3)	1.0×10^{-2} (nd–4.3×10^{-2})	19 (1.0–50)
MM	4.7 (1.9–31)	0.1 (4.5×10^{-2}–0.3)	0.1 (1.7×10^{-2}–0.3)	6.1×10^{-3} (nd–4.9×10^{-2})	24 (13–66)
YD	3.0 (1.4–15)	0.1 (3.8×10^{-2}–1.0)	0.1 (5.3×10^{-2}–8.1)	2.4×10^{-2} (nd–6.7×10^{-2})	13 (6.7–43)
SG	1.1 (0.4–2.8)	6.2×10^{-2} (1.2×10^{-2}–0.5)	0.1 (6.7×10^{-2}–0.3)	nd	9.2 (2.1–30)

[a] Sum of 14 BDE congeners.

[b] Sum of *syn*-DP and *anti*-DP concentrations.

[c] Sum of 60 CB congeners.

[d] Not detectable.

Table S5. Comparisons of concentrations (medians and ranges, ng/g lipid mass) of ∑PBDEs, DBDPE, BTBPE, ∑DP and ∑PCBs in the muscle tissue of waterbirds from various locations around the world.

Species	**Country/site**	**Site description**	**ΣPBDEs**	**DBDPE**	**BTBPE**	***anti*-DP**	***syn*-DP**	**ΣDP**	**ΣPCBs**	**References**
Common kingfisher	China	E-waste site	8.8×10^3	12	7.7					Mo et al., 2012
(*Alcedo atthis*)			(2.0×10^3–2.6×10^4)	(4.5–52)	(2.7–38)					
	China	E-waste site				38	19	58		Mo et al., 2013
						(17–99)	(10–50)	(29–1.5×10^2)		
	China	A nature reserve	89	23	0.2				1.8×10^3	Mo et al., 2013
			(51–4.2×10^2)	(0.4–90)	(0.4×10^{-2}–0.9)				(4.9×10^2–3.0×10^3)	
	China	Reference site				2.7	0.9	3.9		
						(0.8–8.9)	(0.1–3.8)	(1.0–13)		
	China	Reference site	41	4.3	< 0.4×10^{-2}					
			(18–92)	(1.3–9.7)						
Slaty-breasted rail	China	E-waste site	8.2×10^2	22	1.9			1.7×10^2	1.0×10^4	Luo et al., 2009
(*Gallirallus striatus*)			(1.3×10^2–1.4×10^3)	(5-62)	(nd[a]–20)			(14–6.1×10^2)	(4.0×10^3-1.2×10^4)	Zhang et al., 2011a, 2011b
White-breasted waterhen	China	E-waste site	6.0×10^2	13	nd			66	1.8×10^4	
(*Amaurornis phoenicurus*)			(1.5×10^2–1.4×10^4)	(nd–2.8×10^3)	(nd–1.0)			(9–3.6×10^2)	(2.5×10^3–1.4×10^6)	
Ruddy-breasted crake	China	E-waste site	37	10	3.3			56	1.8×10^3	
(*Porzana fusca*)			(23–1.3×10^2)	(4–16)	(nd–9.3)			(21–1.5×10^2)	(9.0×10^2–1.1×10^4)	
Chinese Pond Heron	China	E-waste site	2.2×10^3	1.8×10^2	nd			69	1.2×10^5	
(*Ardeola bacchus*)			(5.3×10^2–2.5×10^3)	(33–8.0×10^2)				(nd–1.3×10^2)	(4.3×10^3–2.7×10^5)	
Commin snipe	China	E-waste site	3.4×10^2	96	nd			7.4	1.0×10^4	
(*Gallinago gallinago*)			(2.7×10^2–1.7×10^3)	(29–1.1×10^2)	(nd–0.9)			(2.3–6.0×10^2)	(8.4×10^3–3.5×10^4)	
Watercock	China	E-waste site	65	12	0.1					Shi et al., 2009
(Gallicrex cinerea)										

Guillemot (*Uria aalge*)	Baltic Sea (Karlsö islands)	Contaminant site	80 [a]	6.8×10^{3} [a]	Lundstedt-Enkel et al., 2006
Black-tailed gull (*Larus crassirostris*)	South Korea	Urbanized coastal region	3.5×10^{3} (7.5×10^{2}–7.1×10^{3})	6.4×10^{4} (9.0×10^{3}–1.3×10^{5})	Hong et al., 2014
Cormorant (*Phalacrocorax carbo*)	Danube Delta, Romania (japsa Marcova)		2.6	7.0×10^{2}	Covaci et al., 2006
	Danube Delta, Romania (channel Papadia)		6.9	2.5×10^{3}	
Fulmar (*Fulmarus glacialis*)	Denmark Faroe Island	Point sources of organohalogen contaminants	16 [b] (11–24)	1.3×10^{4} (1.0×10^{4}–1.6×10^{4})	Fängström et al., 2005
Common cormorant (Phalacrocorax carbo)	Japan (Lake Biwa, Shiga)	/	5.3×10^{2} (2.3×10^{2}–8.2×10^{2})	3.1×10^{6} (6.3×10^{4}–7.7×10^{6})	Kunisue et al., 2008
Yellow-legged gull (*Larus michahellis*)	Turkey (Isikli)	Reference site	47	4.6×10^{2}	Kocagoz et al., 2014
Euroasian coot (*Fulica atra*)			29	21	
Yellow-legged gull (*Larus michahellis*)	Turkey (SÖKE)	Contaminanted by agriculture activites	31 [c]	2.7×10^{2} [c]	
Euroasian coot (*Fulica atra*)			19	36	
Crested kingfisher (*Ceryle rudis*)	South India (Tamil Nadu)			1.6×10^{2} [c, e]	Tanabe et al., 1998

White breasted king-Fisher		40 [c, e]	
(Halcyon smyrnensis)			
Little egret		33 [c, e]	
(Egretta garzetta)			
Pond heron		44 (22–65)	
(*Ardeola grayii*)			
Black-capped kingfishers	North Vietnam	53 (46–60) [d, e]	Minh et al., 2002 [20]
(*Halcyon pileata*)	(Red River estuary)		
Common kingfisher	North Vietnam	59 or 62 [d, e]	
(*Alcedo atthis*)	(Red River estuary)		
common goldeneye	British Columbia,	1.6×10^{3} [c]	Braune et al., 1999
(*Bucephala clangula*)	Canada		[21]
hooded merganser		78 [c]	
(*Lophodytes cucullatus*)			
White-throated kingfisher	North Vietnam	40 [d, e]	
(*Halcyon smyrnensis*)	(Red River estuary)		

[a] Not detectable.

[b] Geometric mean value.

[c] Mean value.

[d] Whole body homogenates.

[e] ng/g wet mass.

Table S6. Median concentrations (ng/g lipid mass).and ranges of PBDEs in the muscle of kingfishers from various locations in a region in South China affected by e-waste recycling.

Compound	E-waste sites	Non–e-waste sites

	QY	GY	ZC	HZ	HY	MZ	JX	JM	MM	YD	ZQ	SG
BDE 28	47 (nd–7.3×10^2)	38 (16–1.5×10^2)	0.5 (0.2–6.1)	0.1 (nd[a]–0.2)	0.1 (0.1–0.3)	0.1 (nd–0.3)	0.4 (0.2–0.8)	0.1 (nd–0.1)	0.1 (nd–0.2)	0.1 (0.1–1.0)	3.7 (1.3–6.3)	0.8 (0.1–2.4)
BDE 47	2.4×10^3 (6–1.6×10^4)	6.3×10^3 (1.7×10^3–1.6×10^4)	32 (21–2.6×10^2)	9.7 (5.9–20)	12 (5.4–19)	5.5 (2.8–12)	29 (19–58)	14 (10–31)	17 (5.3–50)	12 (1.2–30)	8.5 (3.7–24)	1.8 (0.6–4.0)
BDE 99	5.3×10^2 (39–4.6×10^3)	7.3×10^2 (3.7×10^2–6.2×10^4)	3.9 (1.0–39)	2.1 (1.2–2.9)	1.7 (0.5–5.9)	1.0 (0.4–2.8)	1.6 (0.7–17)	2.5 (1.4–4.9)	3.0 (0.5–52)	0.1 (0.04–1.7)	6.5 (1.3–22)	0.8 (0.3–3.4)
BDE 100	8.7×10^2 (1.6×10^2–7.5×10^3)	4.0×10^3(8.4×10^2–8.8×10^3)	15 (3.2–62)	2.0 (0.8–17)	14 (3.3–32)	6.1 (1.7–30)	14 (7.1–59)	17 (4.6–25)	23 (2.5–1.4×10^2)	5.5 (3.2–25)	17 (7.8–62)	7.2 (2.5–23)
BDE 153	1.1×10^3 (1.6×10^2–5.9×10^3)	4.3×10^3 (4.8×10^2–3.2×10^4)	0.4 (0.1–2.4)	16 (4.8–26)	12 (1.2–29)	4.5 (1.9–11)	25 (12–91)	14 (4.1–29)	23 (2.2–83)	7.8 (2.0–27)	14 (5.8–85)	6.4 (3.5–17)
BDE 154	1.4×10^3 (2.5×10^2–1.3×10^4)	2.6×10^3 (5.7×10^2–5.8×10^3)	34 (4.4–1.1×10^2)	59 (11–1.2×10^2)	19 (5.0–40)	6.1 (1.1–12)	17 (8.1–88)	35 (8.2–61)	39 (4.2–1.5×10^2)	9.4 (3.2–42)	25 (12–1.0×10^2)	8.7 (2.7–29)
BDE 183	42 (14–3.6×10^2)	1.7×10^2 (17–7.2×10^2)	3.1 (0.3–10)	3.6 (0.9–13)	2.0 (0.2–3.6)	0.9 (0.4–2.2)	1.8 (0.9–5.6)	2.1 (1.0–3.8)	2.6 (0.6–12)	1.0 (0.2–6.3)	4.0 (1.1–23)	2.0 (0.9–4.4)
BDE 196	1.3×10^2 (58–7.4×10^2)	33 (6.2–1.8×10^2)	2.3 (0.8–16)	0.4 (0.3–2.0)	0.4 (0.3–2.8)	0.3 (0.1–1.6)	0.5 (0.2–1.0)	1.0 (0.3–1.1)	0.3 (0.1–1.5)	1.1 (0.5–3.7)	0.5 (0.3–9.1)	0.3 (0.3–0.7)
BDE 197	2.0×10^2 (74–1.7×10^3)	34 (7.7–3.9×10^2)	0.8 (0.1–3.8)	0.6 (0.4–2.5)	0.8 (0.1–2.2)	0.4 (0.1–0.9)	0.6 (0.2–1.3)	1.2 (0.8–5.0)	1.2 (0.3–2.4)	0.9 (0.5–6.8)	1.5 (0.4–12)	0.6 (0.4–2.0)
BDE 202	5.6×10^2 (1.1×10^2–3.3×10^3)	1.9×10^2 (70–7.6×10^2)	6.8 (1.2–23)	7.2 (1.4–22)	4.9 (2.6–16)	2.2 (1.0–4.2)	4.3 (2.1–18)	0.4 (0.3–0.8)	5.7 (1.7–13)	1.8 (0.7–18)	3.7 (0.8–26)	3.0 (0.8–6.5)
BDE 203	1.0×10^2 (38–7.5×10^2)	28 (5.6–2.0×10^2)	0.7 (0.1–3.5)	0.5 (0.3–1.9)	0.4 (0.1–3.1)	0.3 (0.1–1.4)	0.5 (0.2–1.3)	1.3 (0.4–1.5)	0.6 (0.2–2.0)	0.6 (0.3–5.8)	1.0 (0.4–7.4)	0.4 (0.4–0.9)
BDE 206	3.8×10^2 (1.5×10^2–1.6×10^3)	28 (17–55)	5.2 (2.1–15)	0.5 (0.3–1.1)	0.6 (0.3–1.8)	0.5 (0.1–1.7)	0.6 (0.4–1.4)	1.1 (0.7–1.2)	0.3 (0.2–0.5)	5.0 (0.7–72)	0.9 (0.4–5.1)	0.4 (0.4–3.4)
BDE 207	7.7×10^2 (2.7×10^2–2.9×10^3)	54 (24–3.8×10^2)	5.6 (1.4–27)	1.3 (0.4–2.1)	2.5 (1.3–5.8)	1.3 (0.7–8.0)	2.1 (0.3–5.1)	1.4 (0.6–2.2)	0.9 (0.4–3.0)	3.8 (1.5–18)	2.0 (1.6–9.9)	0.4 (0.4–5.5)
BDE 209	5.0×10^2 (75–5.7×10^3)	1.1×10^2 (25–1.9×10^3)	45 (13–1.0×10^3)	2.0 (0.3–12)	3.8 (0.8–34)	7.6 (1.3–81)	2.2 (0.6–3.5)	1.1 (0.8–2.6)	2.1 (0.1–13)	10 (1.9–41)	3.7 (1.7–25)	1.7 (1.7–11)
ΣPBDEs	9.7×10^3 (2.1×10^3–4.9×10^4)	1.9×10^4 (6.9×10^3–1.3×10^5)	1.6×10^2 (59–1.2×10^3)	1.4×10^2 (46–2.4×10^2)	75 (23–1.4×10^2)	45 (16–1.4×10^2)	1.0×10^2 (64–3.1×10^2)	96 (34–1.7×10^2)	1.1×10^2 (32–4.2×10^2)	70 (25–1.9×10^2)	90 (50–4.1×10^2)	41 (19–92)

[a] Not detectable.

Table S7. Median concentrations (ng/g wet mass).and ranges of PBDEs in the muscle of kingfishers from various locations in a region in South China affected by e-waste recycling.

Compound	E-waste sites		Non–e-waste sites									
	QY	GY	ZC	HZ	HY	MZ	JX	JM	MM	YD	ZQ	SG
BDE 28	1.7 (nd–30)	1.8 (1.0–10)	1.5×10^{-2} (1.2×10^{-2}–7.0×10^{-2})	2.1 (nd[a]–8.1×10^{-3})	8.5×10^{-3} (4.5×10^{-3}–1.1×10^{-2})	2.3×10^{-3} (nd–1.0×10^{-2})	2.0×10^{-2} (1.2×10^{-2}–3.9×10^{-2})	2.1×10^{-3} (nd–5.8×10^{-3})	3.0×10^{-4} (nd–1.4×10^{-2})	4.0×10^{-2} (1.3×10^{-3}–4.1×10^{-2})	0.1 (2.7×10^{-2}–0.2)	1.5×10^{-2} (2.2×10^{-3}–5.7×10^{-2})
BDE 47	99 (3.3–5.3×10^{2})	3.7×10^{2} (1.0×10^{2}–6.3×10^{2})	1.4 (1.0–3.0)	0.4 (0.3–1.0)	0.5 (0.4–0.8)	0.3 (0.2–0.4)	1.5 (1.1–3.2)	0.5 (0.6–1.0)	0.8 (0.3–3.8)	0.5 (6.6×10^{-2}–1.1)	0.2 (0.2–1.2)	3.5×10^{-2} (4.9×10^{-3}–0.1)
BDE 99	21 (2.3–1.6×10^{2})	50 (22–2.5×10^{3})	0.2 (3.8×10^{-2}–0.4)	9.9×10^{-2} (5.0×10^{-2}–0.1)	8.3×10^{-2} (3.4×10^{-2}–0.3)	4.9×10^{-2} (1.6×10^{-2}–0.1)	9.0×10^{-2} (4.5×10^{-2}–0.9)	9.4×10^{-2} (8.9×10^{-2}–0.2)	0.1 (3.2×10^{-2}–3.9)	2.8×10^{-2} (1.9×10^{-3}–6.9×10^{-2})	0.3 (5.7×10^{-2}–0.4)	1.1×10^{-2} (1.7×10^{-4}–0.1)
BDE 100	32 (8.8–2.8×10^{2})	2.4×10^{2}(57–4.0×10^{2})	0.5 (0.1–1.2)	0.7 (0.2–1.2)	0.6 (0.2–2.0)	0.3 (9.8×10^{-2}–1.4)	0.8 (0.4–3.0)	0.7 (0.3–0.8)	1.0 (0.2–10)	0.2 (0.1–1.0)	0.6 (0.3–2.7)	0.2 (2.4×10^{-2}–0.6)
BDE 153	38 (8.9–2.4×10^{2})	2.3×10^{3} (33–1.3×10^{3})	1.3×10^{-2} (5.4×10^{-3}–7.2×10^{-2})	1.1 (0.1–2.7)	0.5 (8.4×10^{-2}–1.7)	0.2 (0.1–0.5)	1.3 (0.6–4.7)	0.9 (0.3–1.0)	0.9 (0.1–4.3)	0.3 (9.0×10^{-2}–1.1)	0.5 (0.3–1.5)	0.2 (2.7×10^{-2}–0.5)
BDE 154	45 (11–4.8×10^{2})	1.2×10^{2} (40–3.9×10^{2})	1.1 (0.2–3.3)	2.4 (0.6–5.4)	0.8 (0.3–2.5)	0.3 (6.4×10^{-2}–0.5)	1.0 (0.4–4.5)	1.6 (0.5–2.0)	1.6 (0.3–7.1)	0.4 (0.1–1.7)	0.8 (0.4–2.5)	0.2 (4.9×10^{-2}–0.7)
BDE 183	2.1 (0.5–14)	8.1 (1.2–37)	0.1 (1.7×10^{-2}–0.3)	0.1 (4.8×10^{-2}–0.5)	7.7×10^{-2} (1.3×10^{-2}–0.2)	4.8×10^{-2} (2.2×10^{-2}–0.1)	9.8×10^{-2} (5.6×10^{-2}–0.2)	0.1 (6.2×10^{-2}–6.1)	0.1 (3.7×10^{-2}–0.6)	3.1×10^{-2} (1.3×10^{-2}–0.2)	0.1 (5.5×10^{-2}–0.3)	4.0×10^{-2} (4.3×10^{-3}–0.1)
BDE 196	5.1 (3.0–23)	1.9 (0.4–7.2)	7.9×10^{-2} (4.2×10^{-2}–0.2)	2.0×10^{-2} (1.4×10^{-2}–6.9×10^{-2})	1.7×10^{-2} (8.9×10^{-3}–0.1)	1.6×10^{-2} (9.2×10^{-3}–5.9×10^{-2})	3.8×10^{-2} (1.4×10^{-2}–5.0×10^{-2})	2.1×10^{-2} (2.1×10^{-2}–4.7×10^{-2})	1.5×10^{-2} (1.6×10^{-3}–7.2×10^{-2})	3.4×10^{-2} (2.2×10^{-2}–0.2)	2.1×10^{-2} (1.4×10^{-2}–0.1)	1.4×10^{-2} (1.5×10^{-3}–2.4×10^{-2})
BDE 197	7.7 (4.4–47)	2.1 (0.5–16)	2.5×10^{-2}	3.3×10^{-2}	4.2×10^{-2}	2.2 (6.9×10^{-2}–	2.8×10^{-2}	3.8×10^{-2}	5.2×10^{-2}	3.1×10^{-2}	4.9×10^{-2}	2.1×10^{-2}

			(1.6×10^{-2}–7.2×10^{-2})	(1.8×10^{-2}–8.6×10^{-2})	(5.3×10^{-2}–8.2×10^{-2})	3.4×10^{-2})	(1.2×10^{-2}–7.2×10^{-2})	(3.8×10^{-2}–0.2)	(1.8×10^{-2}–0.1)	(1.83.1×10^{-2}–0.3)	(2.5×10^{-2}–0.2)	(2.3×10^{-3}–5.4×10^{-2})
BDE 202	21 (4.5–1.5×10^{2})	11 (4.6–31)	0.3 (6.0×10^{-2}–0.9)	0.3 (7.5×10^{-2}–1.1)	0.2 (0.1–0.6)	0.1 (5.8×10^{-2}–0.2)	0.3 (0.1–1.0)	0.1 (1.6×10^{-2}–2.5×10^{-2})	0.2 (0.1–0.6)	7.4×10^{-2} (2.8×10^{-2}–0.7)	0.1 (2.5×10^{-2}–0.8)	4.3×10^{-2} (6.3×10^{-3}–0.2)
BDE 203	3.6 (1.6–35)	1.7 (0.4–8.2)	2.6×10^{-2} (2.5×10^{-3}–7.2×10^{-2})	2.2×10^{-2} (1.4×10^{-2}–6.5×10^{-2})	1.9×10^{-2} (1.2×10^{-3}–0.1)	1.5×10^{-2} (7.8×10^{-3}–5.3×10^{-2})	2.5×10^{-2} (1.5×10^{-2}–7.4×10^{-2})	2.7×10^{-2} (2.7×10^{-2}–5.9×10^{-2})	1.9×10^{-2} (7.7×10^{-3}–0.1)	2.3×10^{-2} (1.0×10^{-2}–0.2)	3.3×10^{-2} (1.5×10^{-2}–0.1)	1.4×10^{-2} (1.7×10^{-3}–2.1×10^{-2})
BDE 206	14 (6.3–61)	1.4 (1.1–3.4)	0.2 (0.1–0.6)	2.7×10^{-2} (1.2×10^{-2}–3.8×10^{-2})	3.1×10^{-2} (1.6×10^{-2}–6.8×10^{-2})	2.5×10^{-2} (3.3×10^{-3}–6.3×10^{-2})	2.7×10^{-2} (1.8×10^{-2}–7.8×10^{-2})	2.7×10^{-2} (3.9×10^{-2}–5.2×10^{-2})	3.1×10^{-2} (1.6×10^{-2}–6.8×10^{-2})	0.2 (4.4×10^{-2}–2.4)	2.7×10^{-2} (1.6×10^{-2}–6.8×10^{-2})	1.1×10^{-2} (4.2×10^{-3}–0.1)
BDE 207	30 (13–65)	3.5 (1.6–15)	0.2 (0.1–1.3)	6.5×10^{-2} (2.2×10^{-2}–0.1)	0.1 (7.2×10^{-2}–0.2)	6.2×10^{-2} (3.2×10^{-2}–0.4)	0.1 (1.5×10^{-2}–0.2)	6.2×10^{-2} (4.0×10^{-2}–7.3×10^{-2})	0.1 (7.2×10^{-2}–0.2)	0.1 (7.8×10^{-2}–0.7)	8.6×10^{-2} (4.7×10^{-2}–0.1)	2.3×10^{-2} (3.0×10^{-3}–0.2)
BDE 209	7.9 (4.0–1.5×10^{2})	6.2 (1.7–74)	1.2 (0.7–52)	8.3×10^{-2} (1.4×10^{-2}–0.6)	0.2 (3.7×10^{-2}–1.3)	0.4 (5.9×10^{-2}–3.0)	0.1 (3.0×10^{-2}–0.2)	8.3×10^{-2} (4.7×10^{-2}–8.6×10^{-2})	0.2 (3.7×10^{-2}–1.3)	0.5 (0.2–8.7)	0.2 (5.2×10^{-2}–0.4)	0.2 (2.2×10^{-2}–0.4)
ΣPBDEs	3.5×10^{2} (1.2×10^{3}–1.7×10^{3})	1.0×10^{3} (3.5×10^{2}–5.1×10^{3})	5.2 (3.1–60)	5.4 (2.4–11)	3.5 (1.6–8.2)	2.0 (1.1–5.2)	5.5 (3.4–16)	4.4 (2.1–5.5)	4.7 (1.9–31)	2.6 (1.4–16)	3.0 (2.2–9.0)	1.1 (0.4–2.8)

[a] Not detectable

Table S8. Median concentrations (ng/g lipid mass).and ranges of AHFRs in the muscle of kingfishers from different sampling sites.

Locations	ΣAHFRs [a]	DBDPE	BTBPE	*syn*-DP	*anti*-DP	ΣDP [b]
E-waste sites						
QY	61 (8.5–3.6×10^2)	2.4 (0.3–1.1×10^2)	0.6 (nd [c] –49)	14 (4.4–66)	40 (8.6–1.7×10^2)	57 (13–2.4×10^2)
GY	2.0×10^2 (45–2.9×10^2)	25 (20–39)	9.0 (5.6–12)	70 (2.5–1.1×10^2)	1.0×10^2 (16–1.6×10^2)	1.7×10^2 (19–2.2×10^2)
Non–e-waste sites						
ZQ	33 (22–94)	23 (0.4–90)	0.2 (0.04–0.4)	0.7 (0.09–2.4)	2.7 (0.4–6.6)	3.9 (1.0–13)
ZC	20 (5.8–2.9×10^2)	18 (4–2.3×10^2)	0.2 (nd–0.5)	0.7 (0.2–15)	3.9 (1.0–50)	4.8 (1.2–65)
MZ	6.4 (2.2–47)	4.0 (1.6–34)	0.1 (0.02–0.7)	0.3 (0.09–4.0)	0.9 (0.3–13)	1.2 (0.4–17)
JX	3.7 (0.8–9.5)	2.7 (0.01–8.7)	0.1 (nd–0.2)	0.08 (nd–0.6)	0.9 (0.4–2.2)	0.9 (0.4–2.5)
HY	5.7 (1.3–33)	3.2 (2.3–13)	0.1 (nd–0.4)	0.5 (0.1–1.8)	1.7 (0.5–4.7)	2.2 (0.7–6.5)
HZ	5.6 (2.3–20)	3.9 (1.1–15)	0.1 (0.02–1.3)	0.5 (0.2–1.0)	2.0 (0.7–4.7)	2.2 (0.9–5.7)
SG	7.2 (3.6–22)	4.3 (1.9–9.7)	nd	0.5 (0.2–2.3)	1.6 (0.6–11)	2.2 (0.8–12)
YD	7.9 (2.0–2.3×10^2)	3.9 (1.2–24)	0.2 (nd–1.7)	0.9 (0.03–5.4)	2.6 (0.6–19)	2.8 (0.7–24)
JM	4.8 (2.4–13)	1.6 (1.6–8.2)	0.06 (nd–0.9)	0.4 (0.2–0.6)	1.6 (0.5–4.3)	2.2 (0.7–4.7)
MM	5.1 (2.4–7.2)	3.2 (0.8–4.4)	0.2 (nd–1.0)	0.5 (0.2–0.9)	1.6 (0.4–2.7)	2.0 (0.7–3.6)

[a] ΣAHFRs were sum of concentrations of DBDPE, BTBPE and DP.

[b] sum of *syn*-DP and *anti*-DP concentrations.

[c] Not detectable.

Table S9. Hazard quotients (HQs) of PCBs and PBDEs for kingfishers.

Location	PCBs						PBDEs		
	HQ_{10} Mean	HQ_{10} Min	HQ_{10} Max	HQ_{50} Mean	HQ_{50} Min	HQ_{50} Max	HQ_{05} Mean	HQ_{05} Min	HQ_{05} Max
E-waste sites									
QY	33	1.9	1.5×10^{2}	2.3	0.1	10	24	5.4	77
GY	2.2	0.4	6.3	0.2	2.8×10^{-2}	0.4	73	16	2.2×10^{2}
Non–e-waste sites									
ZQ	0.1	5.9×10^{-2}	0.2	0.8×10^{-2}	0.4×10^{-2}	1.2×10^{-2}	0.2	9.5×10^{-2}	0.4
ZC	5.6×10^{-2}	0.7×10^{-2}	0.2	0.4×10^{-2}	0.1×10^{-2}	1.5×10^{-2}	0.5	0.1	2.7
MZ	1.4×10^{-2}	0.6×10^{-2}	2.5×10^{-2}	0.1×10^{-2}	0.4×10^{-3}	0.2×10^{-2}	9.9×10^{-2}	5.0×10^{-2}	0.2
JX	2.4×10^{-2}	0.9×10^{-2}	4.7×10^{-2}	0.2×10^{-2}	0.1×10^{-2}	0.3×10^{-2}	0.3	0.1	0.7
HY	2.3×10^{-2}	0.6×10^{-2}	4.3×10^{-2}	0.2×10^{-2}	0.4×10^{-3}	0.3×10^{-2}	9.9×10^{-2}	5.0×10^{-2}	0.2
HZ	3.7×10^{-2}	1.5×10^{-2}	0.1	0.3×10^{-2}	0.1×10^{-2}	0.8×10^{-2}	0.3	0.1	0.5
SG	5.3×10^{-2}	2.1×10^{-2}	0.1	0.4×10^{-2}	0.1×10^{-2}	0.8×10^{-2}	6.0×10^{-2}	2.2×10^{-2}	0.1
YD	5.5×10^{-2}	2.2×10^{-2}	0.2	0.4×10^{-2}	0.2×10^{-2}	1.1×10^{-2}	0.2	6.0×10^{-2}	0.7
JM	7.9×10^{-2}	3.2×10^{-2}	0.1	0.5×10^{-2}	0.2×10^{-2}	1.0×10^{-2}	0.2	9.4×10^{-2}	0.2
MM	8.3×10^{-2}	3.8×10^{-2}	0.1	0.6×10^{-2}	0.3×10^{-2}	1.0×10^{-2}	0.2	0.2	0.4

Table S10. Toxic equivalent quantity (TEQ) concentrations (pg/g wet mass) of major coplanar PCB in kingfishers from a region in South China affected by recycling of electronic waste.

Locations	TEQs [a]		
	Mean	Median	Range
E-waste sites			
QY	1.3×10^3	7.2×10^2	1.7×10^2–4.9×10^3
GY	4.8	3.5	0.9–13
Non–e-waste sites			
ZC	23	13	10–63
ZQ	36	34	21–62
HZ	23	15	8.4–48
JX	14	13	5.1–32
MZ	5.8	3.9	1.9–17
HY	9.8	9.3	3.2–19
JM	39	29	18–69
MM	46	33	10–1.4×10^2
YD	7.4	7.2	0.1–17
SG	31	25	5.9–72

[a] The TEQ concentrations were estimated using the amounts of each PCB congener multiplied with their toxic equivalency factor for birds ($TEF_{WHO\text{-}Avian}$) proposed by World Health Organization.

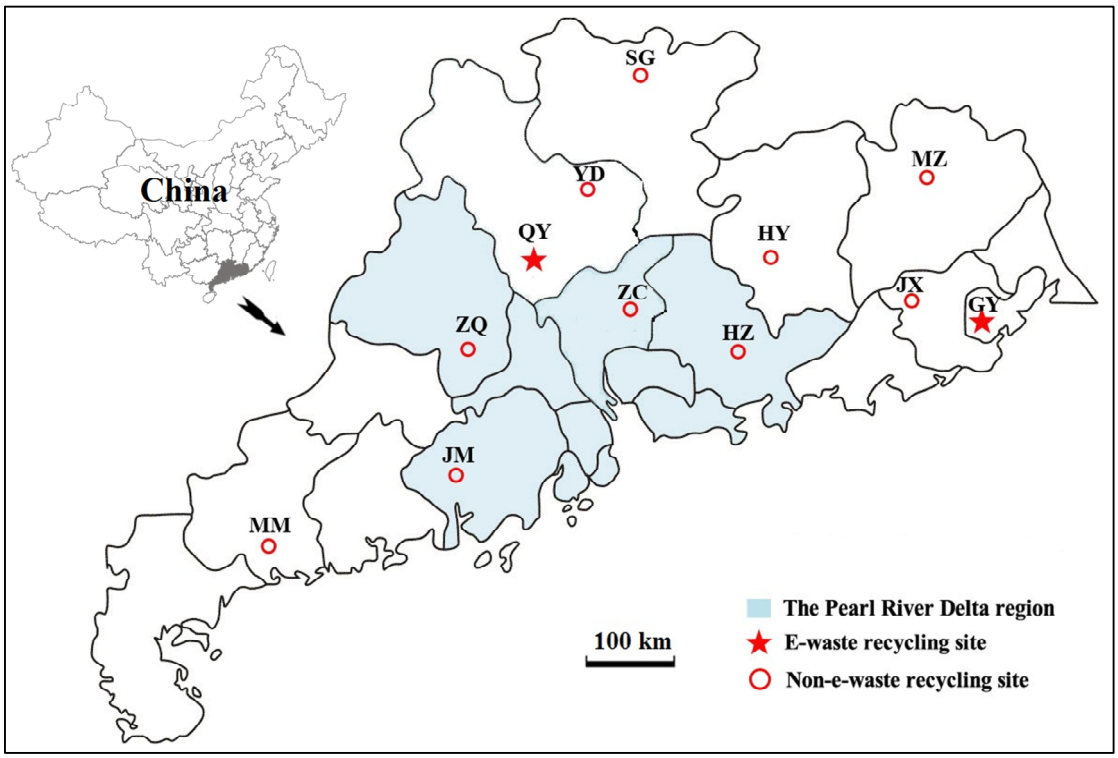
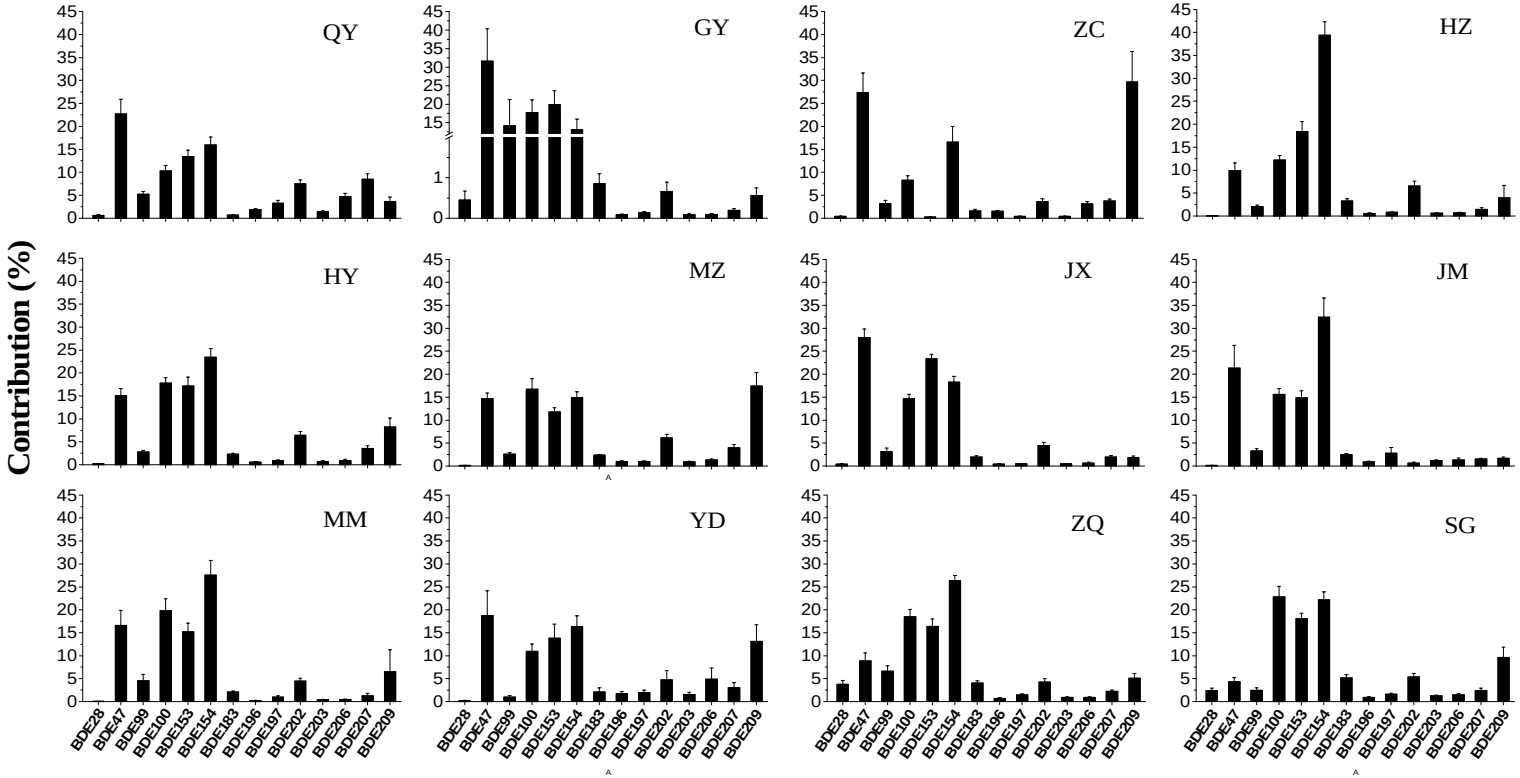


Figure S1. Sampling locations.

Figure S2. Profiles of PBDE congeners in the muscle of kingfishers from 12 locations in a region of South China affected by e-waste recycling.



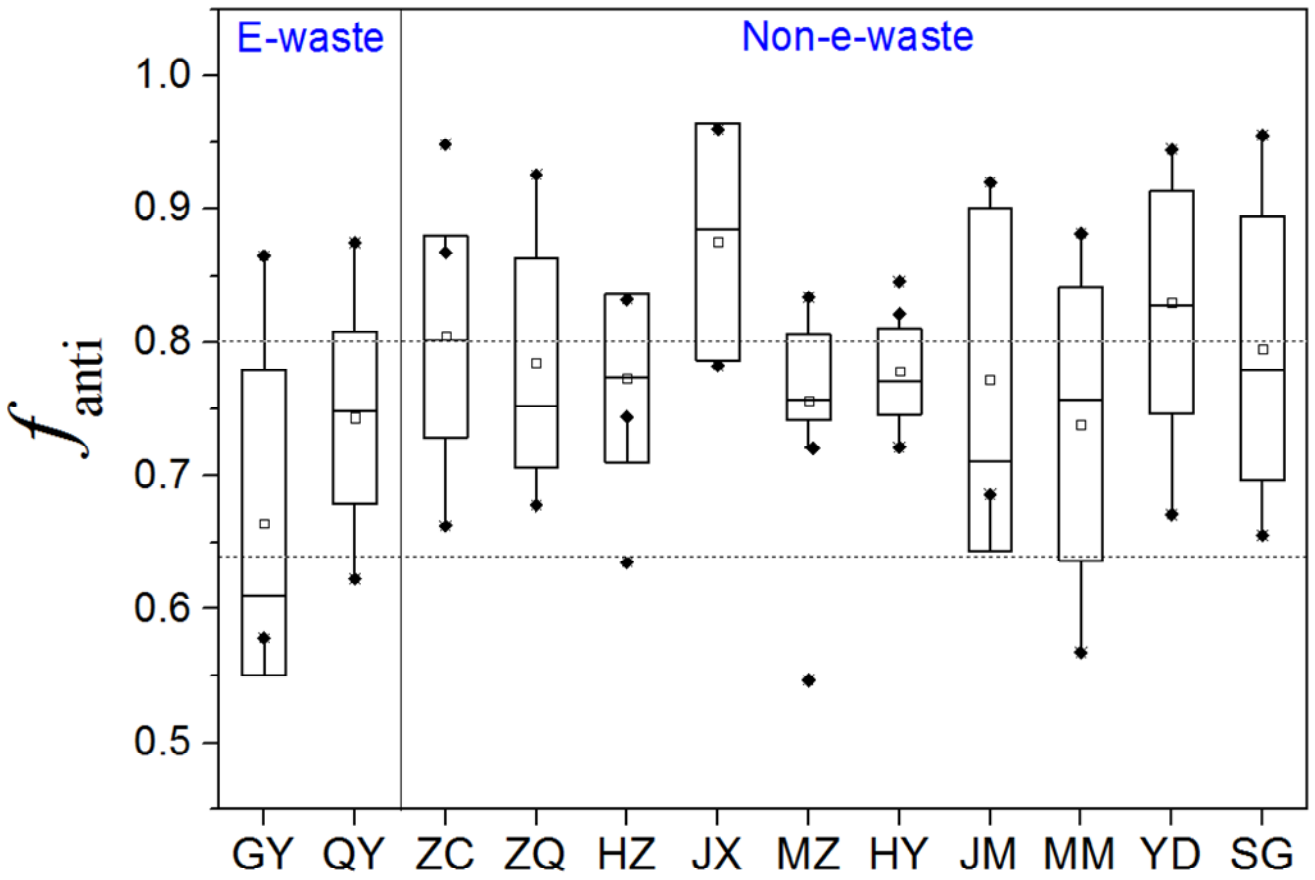


Figure S3. Values of f_{anti} in muscle of kingfishers.

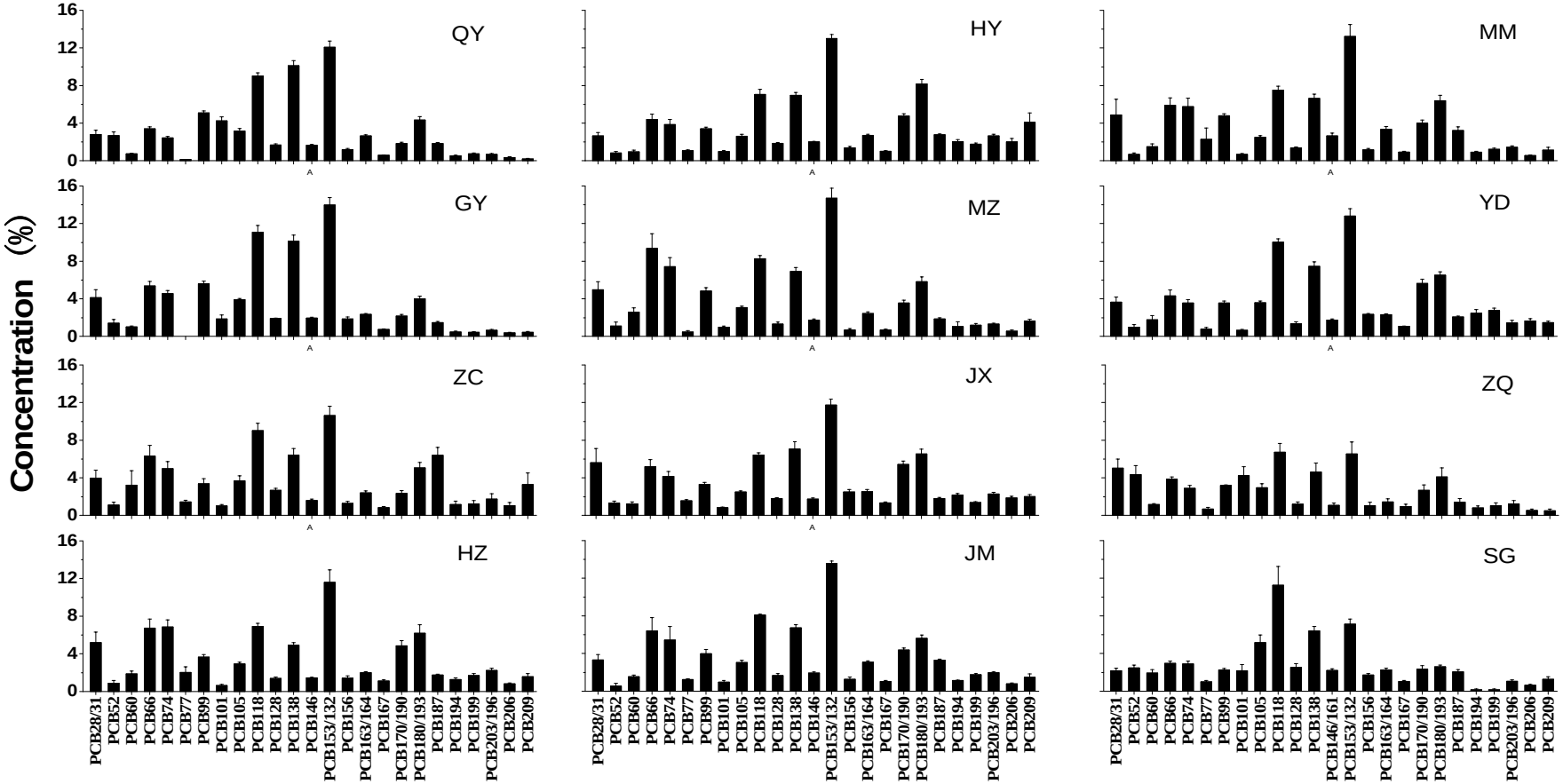


Figure S4. Profiles of PCB congeners in the muscle of kingfishers.

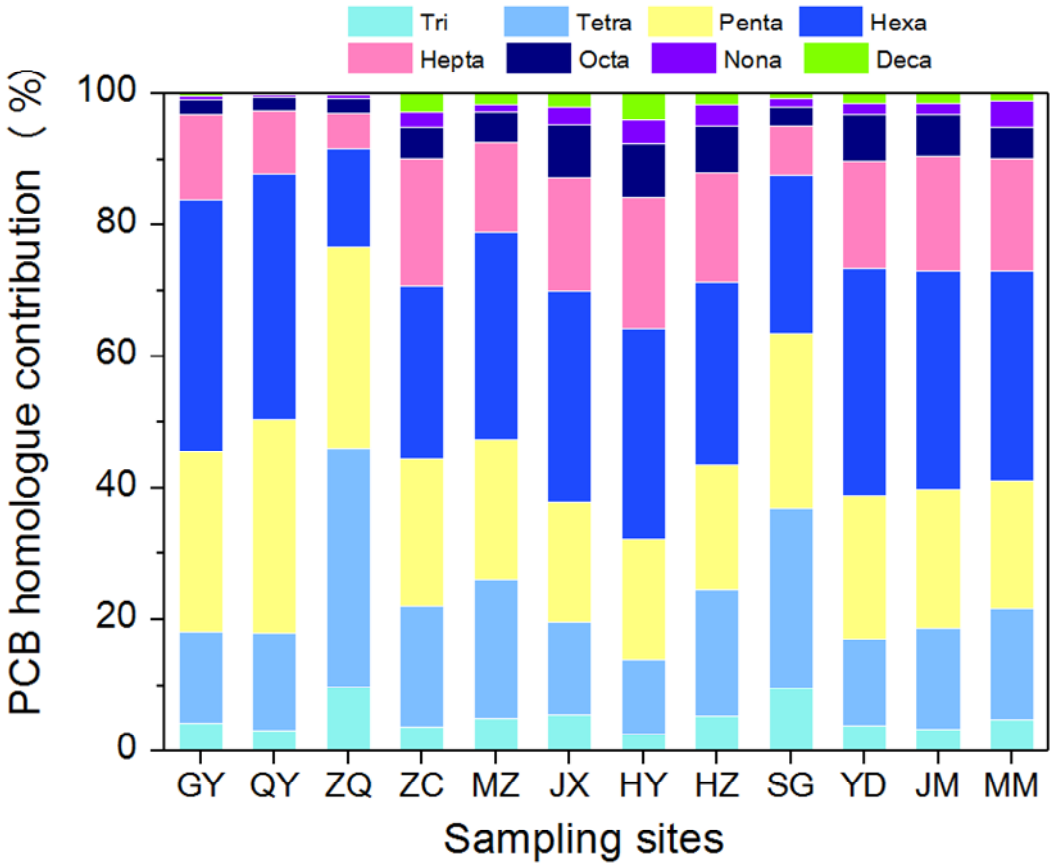


Figure S5. PCB homologue profiles in kingfishers.

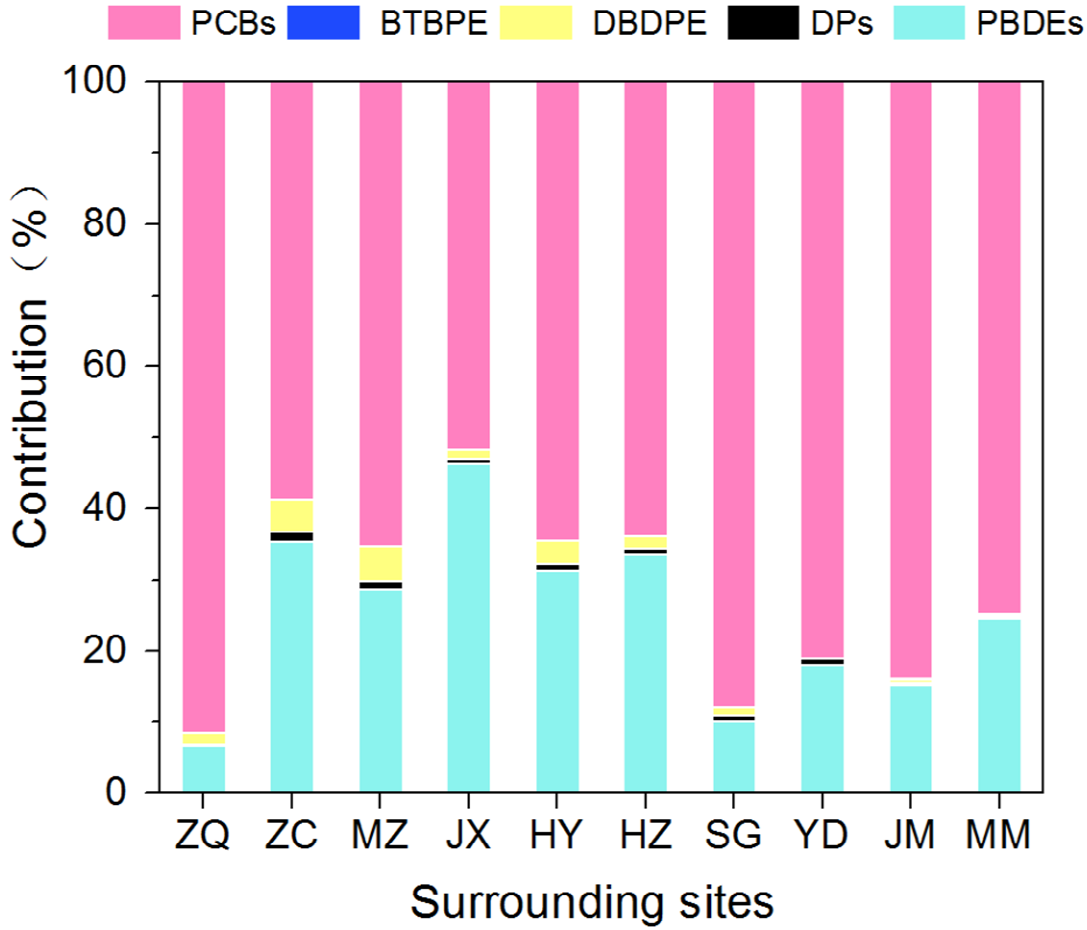


Figure S6. Compositional profiles for PBDEs, AHFRs (including DP, DBDPE and BTBPE) and PCBs in kingfishers collected from non–e-waste sites.

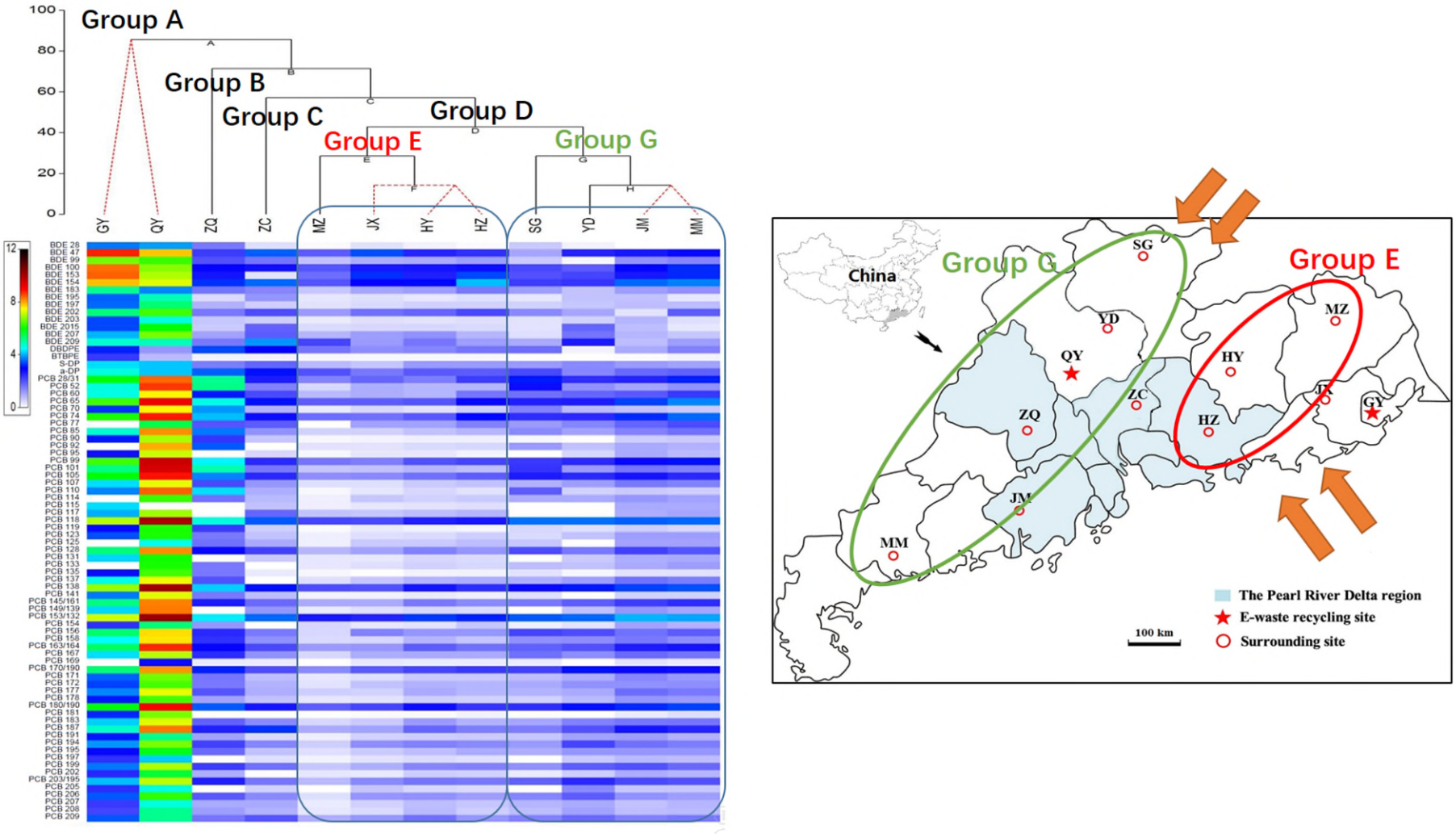


Figure S7. Grouping map

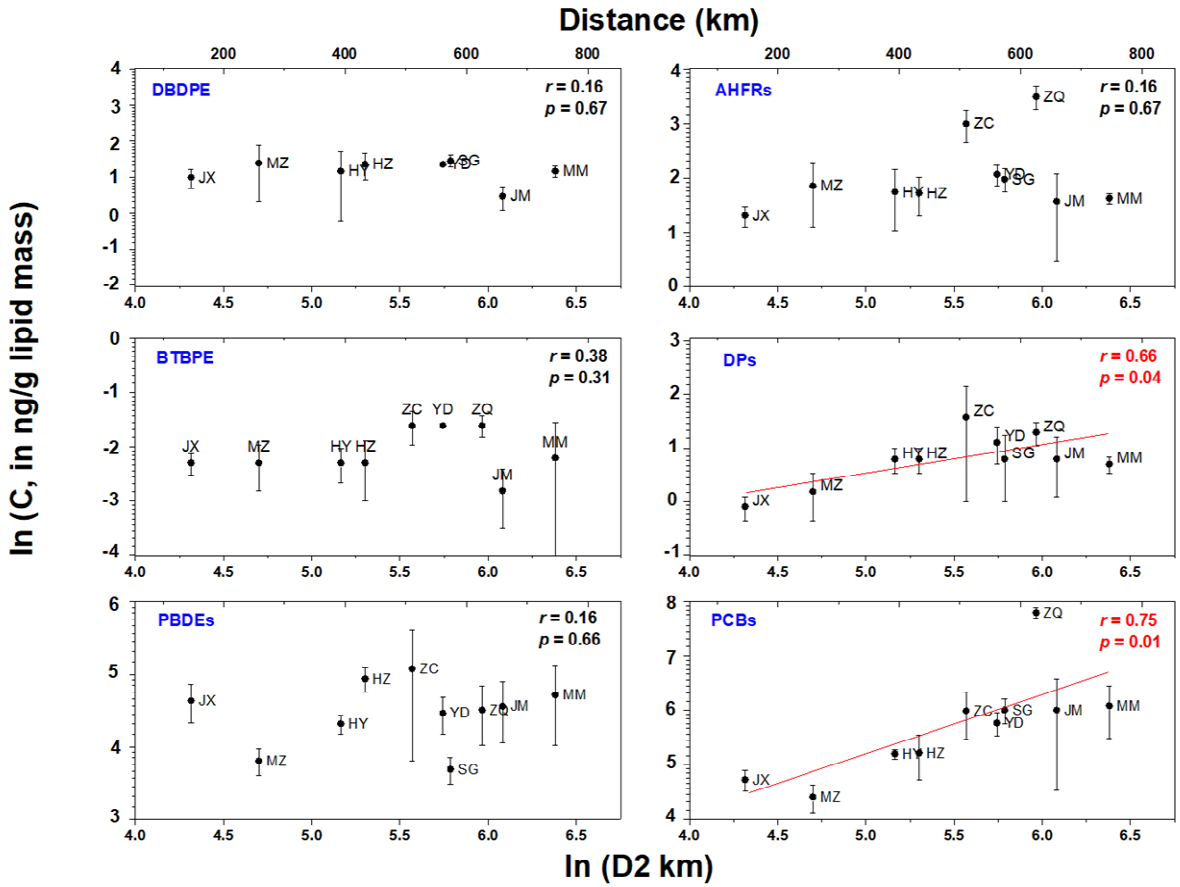


Figure S8. Correlations analysis between the natural logarithm (In) concentrations (ng/g lipid mass) of HFRs (including PBDEs, DP, DBDPE and BTBPE) and PCBs and the natural logarithm distance (75–440 km) from e-waste recycling site GY (In D2). (Top x-axis: distance; Bottom x-axis: logarithm distance)

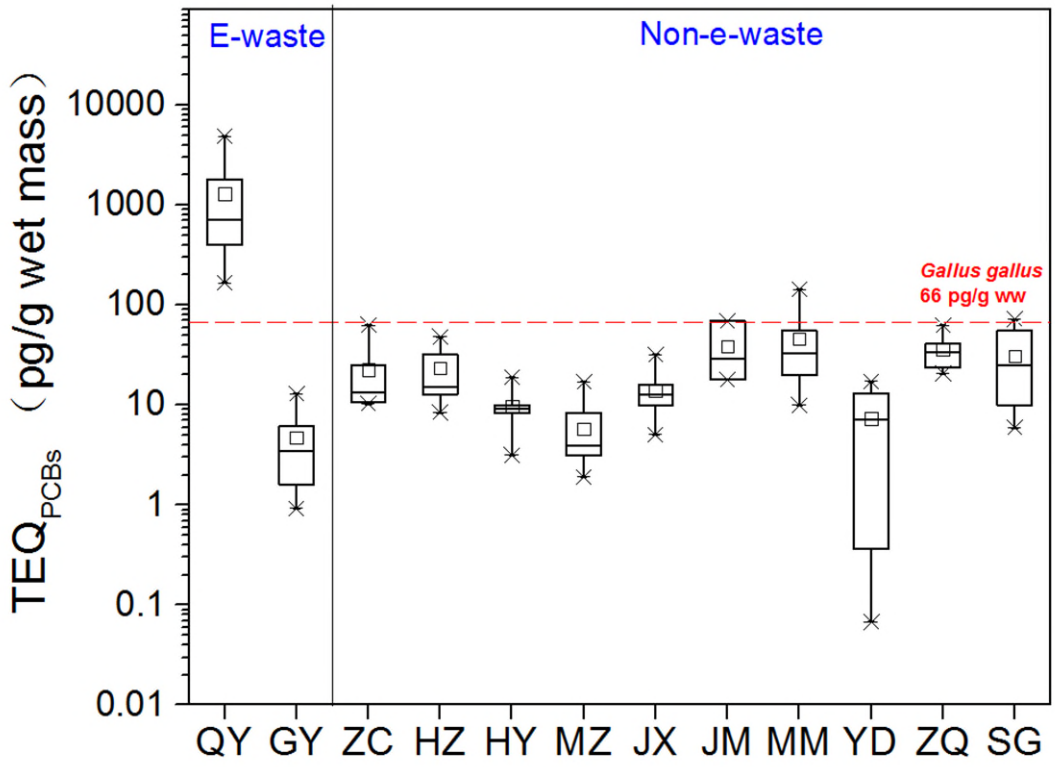


Figure S9. Toxic equivalent quantity (TEQ) concentrations of major coplanar PCB (pg/g wet mass) in kingfishers from a region in South China affected by e-waste recycling. Box plots are defined as follows: center line, median value; hallow square, mean value; box plot edges, the 25th and 75th centile.

References

Braune, B., Malone, B., Burgess, N., Elliott, J., Garrity, N., Hawkings, J., Hines, J., Marshall, H., Marshall, W., Rodrigue, J., 1999. Chemical residues in waterfowl and gamebirds harvested in Canada, 1987–95. Technical Report Series No. 326. Canadian Wildlife Service, Ottawa.

Covaci, A., Gheorghe, A., Hulea, O., Schepens, P., 2006. Levels and distribution of organochlorine pesticides, polychlorinated biphenyls and polybrominated diphenyl ethers in sediments and biota from the Danube Delta, Romania. Environ. Pollut. 140, 136–149.

Fängström, B., Athanasiadou, M., Athanassiadis, I., Bignert, A., Grandjean, P., Weihe, P., Bergman, Å., 2005. Polybrominated diphenyl ethers and traditional organochlorine pollutants in fulmars (*Fulmarus glacialis*) from the Faroe Islands. Chemosphere. 60, 836–843.

Fernie, K. J., Shutt, J. L., Letcher, R. J., Ritchie, I. J., Bird, D. M., 2009. Environmentally relevant concentrations of DE-71 and HBCD alter eggshell thickness and reproductive success of American kestrels. Environ. Sci. Technol. 43, 2124–2130.

Gilchrist, T. T., Letcher, R. J., Thomas, P., Fernie, K. J., 2014. Polybrominated diphenyl ethers and multiple stressors influence the reproduction of free-ranging tree swallows (*Tachycineta bicolor*) nesting at wastewater treatment plants. Sci. Total Environ. 472, 63–71.

Henny, C. J., Kaiser, J. L., Grove, R. A., Johnson, B. L., Letcher, R. J., 2009. Polybrominated diphenyl ether flame retardants in eggs may reduce reproductive success of ospreys in Oregon and Washington, USA. Ecotoxicology. 18, 802–813.

Hong, S. H., Shim, W. J., Han, G. M., Ha, S. Y., Jang, M., Rani, M., Hong, S., Yeo, G. Y., 2014. Levels and profiles of persistent organic pollutants in resident and migratory birds from an urbanized coastal region of South Korea. Sci. Total Environ. 470, 1463–1470.

Kunisue, T., Higaki, Y., Isobe, T., Takahashi, S., Subramanian, A., Tanabe, S., 2008. Spatial trends of polybrominated diphenyl ethers in avian species: Utilization of stored samples in the environmental specimen bank of Ehime University (es-Bank). Environ. Pollut. 154, 272–282.

Kocagoz, R., Onmus, O., Onat, I., Cagdas, B., Siki, M., 2014. Orhan, H., Environmental and biological monitoring of persistent organic pollutants in waterbirds by non-invasive versus invasive sampling. Toxicol Lett. 230, 208–217.

Lundstedt-Enkel, K., Asplund, L., Nylund, K., Bignert, A., Tysklind, M., Olsson, M., Orberg, J., 2006. Multivariate data analysis of organochlorines and brominated flame retardants in Baltic Sea guillemot (*Uria aalge*) egg and muscle. Chemosphere. 65, 1591–1599.

Luo, X. J., Zhang, X. L., Liu, J., Wu, J. P., Luo, Y., Chen, S. J., Mai, B. X., Yang, Z. Y., 2009. Persistent halogenated compounds in waterbirds from an e-waste recycling region in South China. Environ Sci Technol. 43, 306–11.

McKernan, M. A., Rattner, B. A., Hale, R. C., Ottinger, M. A., 2009. Toxicity of polybrominated diphenyl ethers (De-71) in chicken (*Gallus Gallus*), mallard (*Anas Platyrhynchos*), and American kestrel (*Falco Sparverius*) embryos and hatchlings. Environ Toxicol Chem. 28, 1007–1017.

Minh, T. B., Kunisue, T., Yen, N. T. H., Watanabe, M., Tanabe, S., Hue, N. D., Qui, V., 2001. Persistent organochlorine residues and their bioaccumulation profiles in resident and migratory birds from North Vietnam. Environ. Toxicol. Chem. 21, 2108–2118.

Mo, L., Wu, J. P., Luo, X. J., Zou, F. S., Mai, B. X., 2012. Bioaccumulation of polybrominated diphenyl ethers, decabromodiphenyl ethane, and 1,2-bis(2,4,6-tribromophenoxy) ethane flame retardants in kingfishers (*Alcedo atthis*) from an electronic waste-recycling site in South China. Environ Toxicol Chem. 31, 2153–2158.

Mo, L., Wu, J. P., Luo, X. J., Li, K. L., Peng, Y., Feng, A. H., Zhang, Q., Zou, F. S., Mai, B. X., 2013. Using the kingfisher (*Alcedo atthis*) as a bioindicator of PCBs and PBDEs in the dinghushan biosphere reserve, China. Environ Toxicol Chem. 32, 1655–1662.

Peng, Y., Wu, J. P., Tao, L., Mo, L., Tang, B., Zhang, Q., Luo, X. J., Zou, F. S., Mai, B. X., 2015. Contaminants of legacy and emerging concern in terrestrial passerines from a nature reserve in South China: Residue levels and inter-species differences in the accumulation. Environ. Pollut. 203, 7–14.

Shi, T., Chen, S.J., Luo, X.J., Zhang, X.L., Tang, C.M., Luo, Y., Ma, Y.J., Wu, J.P., Peng, X.Z., Mai, B.X., 2009. Occurrence of brominated flame retardants other than polybrominated diphenyl ethers in environmental and biota samples from southern China. Chemosphere. 74, 910–916.

Tanabe, S., Senthilkumar, K., Kannan, K., Subramanian, A. N., 1998. Accumulation features of polychlorinated biphenyls and organochlorine pesticides in resident and migratory birds from South India. Arch Environ Con Tox. 34, 387–397

Van den Steen, E., Eens, M., Jaspers, V. L. B., Covaci, A., Pinxten, R., 2009. Effects of laying order and experimentally increased egg production on organic pollutants in eggs of a terrestrial songbird species, the great tit (*Parus major*). Sci. Total Environ. 407, 4764–4770.

Zhang, X. L., Luo, X. J., Liu, H. Y., Yu, L. H., Chen, S. J., Mai, B. X., 2011a. Bioaccumulation of several brominated flame retardants and dechlorane plus in waterbirds from an e-waste recycling region in South China: Associated with trophic level and diet sources. Environ. Sci. Technol. 45, 400–405.

Zhang, X. L., Luo, X. J., Liu, J., Luo, Y., Chen, S. J., Mai, B. X., 2011b. Polychlorinated biphenyls and organochlorinated pesticides in birds from a contaminated region in South China: association with trophic level, tissue distribution and risk assessment. Environ Sci Pollut Res. 18, 556–565.