



Species-specific biomagnification and habitat-dependent trophic transfer of halogenated organic pollutants in insect-dominated food webs from an e-waste recycling site

Yu Liu^{a,b}, Xiaojun Luo^{a,*}, Yanghong Zeng^a, Wenqing Tu^b, Mi Deng^b, Yongming Wu^{b,*}, Bixian Mai^a

^a State Key Laboratory of Organic Geochemistry and Guangdong Key Laboratory of Environmental Protection and Resources Utilization, Guangzhou Institute of Geochemistry, Chinese Academy of Sciences, Guangzhou 510640, People's Republic of China

^b Research Institute of Poyang Lake, Jiangxi Academy of Sciences, Nanchang 330012, People's Republic of China

ARTICLE INFO

Handling Editor: Adrian Covaci

Keywords:

Halogenated organic pollutants
Biomagnification factor
Trophic magnification factor
Insect-dominated food web
E-waste

ABSTRACT

Aquatic, amphibious, and terrestrial organisms in or around a pond that was contaminated by e-waste were collected and persistent halogenated organic pollutants (HOPs) for these species were analyzed. Based on the stable isotope and dietary composition, the aquatic and terrestrial food webs and several insect-dominated food chains including insects – toads, insects – lizards, and insects – birds were constructed. Biomagnification factors (BMFs) for insect-dominated food chains and trophic magnification factors (TMFs) in aquatic and terrestrial food webs were calculated. The BMFs of HOPs (except DBDPE) in insect – bird food chains were significantly higher than those in insect – toad and insect – lizard food chains, indicating that HOPs accumulated more easily in homeotherms than in poikilotherms. Trophic magnification was present for most of the PCB congeners in both aquatic and terrestrial food webs. Differences between the trophic transfer of halogenated flame retardant in terrestrial and aquatic food webs were observed, with trophic magnification in the terrestrial food web but trophic dilution in the aquatic food web for most of chemicals (except for lower brominated PBDE congeners). Meanwhile, the contour plots of TMFs across combinations of $\log K_{OW}$ and $\log K_{OA}$ for terrestrial food web were distinct from those for aquatic food web. These results indicate that the biomagnification mechanisms of HOPs in aquatic food webs are different from those in terrestrial food webs, and further suggest that the bioaccumulation of contaminants in terrestrial ecosystems cannot be directly deduced from aquatic ecosystems.

1. Introduction

Halogenated organic pollutants (HOPs), such as polychlorinated biphenyls (PCBs), polybrominated diphenyl ethers (PBDEs), and polybrominated biphenyls (PBBs), are ubiquitous contaminants in the environment, particularly in typically contaminated areas (Jepson and Law, 2016). Due to the regulation of the production and usage of PCBs, PBDEs, and PBBs; some alternative halogenated flame retardants (HFRs), including dechlorane plus (DP), pentabromotoluene (PBT), pentabromoethylbenzene (PBEB), hexabromobenzene (HBB), and decabromodiphenyl ethane (DBDPE) have been used as alternatives for certain applications (Covaci et al., 2011). However, these alternative HFRs have similar physicochemical properties to those of traditional HOPs and available data have indicated that they may be prone to bioaccumulation and biomagnification in food webs, resulting in significant environmental risks (Baron et al., 2015; Tao et al., 2019).

Biomagnification potential is one of the vital criteria for assessing

the ecological risks of chemicals, particularly those of persistent organic pollutants (Gobas et al., 2016). The biomagnification factors (BMFs) and trophic magnification factors (TMFs) of HOPs have been widely studied in aquatic and terrestrial food webs (Voorspoels et al., 2007; Yu et al., 2013; Walters et al., 2016; Morris et al., 2018). A few studies have identified differences in biomagnification potentials of HOPs (particularly PBDEs) between aquatic and terrestrial ecosystems (Chen and Hale, 2010; Liu et al., 2020). Limited studies on PBDEs in the terrestrial food webs have suggested that unlike the aquatic food webs, stronger biomagnification potential for high brominated congeners (octa- to deca-BDEs) can be observed in terrestrial predators (Park et al., 2009; Yu et al., 2011; Morris et al., 2018). Kelly et al. (2007) constructed a numerical model and reported that organic contaminants with intermediate to high octanol–water partition coefficients ($K_{OW} > 10^2$) and octanol–air partition coefficients ($K_{OA} > 10^6$), can theoretically be biomagnified more effectively in terrestrial food chains than in aquatic ones. However, the biomagnification potential of

* Corresponding authors.

E-mail addresses: luoxiao@igc.ac.cn (X. Luo), wuyongming@jxas.ac.cn (Y. Wu).

<https://doi.org/10.1016/j.envint.2020.105674>

Received 19 December 2019; Received in revised form 17 March 2020; Accepted 17 March 2020

Available online 28 March 2020

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contaminants in terrestrial environments has only been reported using simple models or used data collected from a limited number of trophic levels or species groups (Kelly and Gobas, 2001, 2003). Thus, there is a critical need for more terrestrial field studies that assess biomagnification and provide essential empirical field data on organic chemicals, especially emerging contaminants (Gobas et al., 2016).

As mentioned above, most studies on the biomagnification of HOPs in terrestrial food webs have focused on high trophic level predators, such as raptors and mammals (Voorspoels et al., 2007; Yu et al., 2011; Morris et al., 2018), but have largely ignored the role played by insects. Insects are an important link between primary producers (plants) and secondary consumers, and play an important role in accumulating and transferring pollutants to insectivores, particularly in terrestrial ecosystems (Rosenberg et al., 1986). A few studies have calculated the BMF values of HOPs in the insect-dominated terrestrial food chains. For example, Blankenship et al. (2005) reported that the BMF values of PCBs in the terrestrial invertebrates (*Coleoptera*) – predators (shrew, house wren, and bluebird) food chain range from 0.83 to 35. Nie et al. (2015) found that the BMFs of PBDEs in the insects (grasshopper, ant, butterfly, and dragonfly) – turtledoves food chain range from 0.20 to 11.6. Only one study has indicated that the TMF values of HOPs in the terrestrial food web (mainly insects-birds) ranged from 1.20 to 15.66 (Fremlin, 2018). However, the number of species and sample sizes of insects were relatively small in the studies above, and some insects were tested in mixed samples or not clearly identified. Therefore, further comprehensive and systematic study of the insect-dominated food chains is necessary.

E-waste is a critical global environmental health issue, particularly in developing countries (Zhang et al., 2012). In the previous study, we reported the levels and profiles of HOPs in insects and their predators from an e-waste recycling site in South China (Liu et al., 2018a; 2018b). However, we did not address the biomagnification magnitudes of HOPs in the insect-dominated food chains in that study. In this study, trophic levels (TLs) based on the $\delta^{15}\text{N}$ of different species were determined and the biomagnification of HOPs was assessed by calculating their BMFs and TMFs. Our main objective is to contribute datasets for HOPs biomagnification in terrestrial food webs. Our second objective is to conduct a comparative analysis between the TMF values of aquatic and terrestrial food webs, and to demonstrate the effects of partition coefficients (K_{OW} and K_{OA}) on the biomagnification of HOPs.

2. Materials and methods

2.1. Sampling

Samples were collected from a pond and the surrounding region (within a 500 m radius) located in Longtang Town, Qingyuan County of Guangdong Province. The pond was heavily polluted by chemicals associated with e-waste that had been discarded there. Details regarding the sampling site and sampling methods were provided in our previous paper (Liu et al., 2018a; 2018b). Six orders of insects, namely *Odonata* ($n = 15$), *Coleoptera* ($n = 12$), *Lepidoptera* ($n = 21$), *Orthoptera* ($n = 31$), *Hemiptera* ($n = 25$), and *Mantodea* ($n = 3$), were collected between September of 2015 and November of 2016. At the same time, we also collected aquatic, amphibious, and terrestrial predators, including fish (*Cyprinus carpio*, $n = 6$), prawn (*Macrobrachium nipponense*, $n = 5$), watersnake (*Enhydris chinensis*, $n = 7$), White-breasted waterhen egg (*Amaurornis phoenicurus*, $n = 6$), frog (*Kaloula pulchra*, $n = 5$), toad (*Duttaphrynus melanostictus*, $n = 6$), lizard (*Calotes versicolor*, $n = 10$), Eurasian blackbird (*Turdus merula*, $n = 2$), Oriental magpie-robin (*Copsychus saularis*, $n = 3$), and Long-tailed shrike (*Lanius schach*, $n = 2$). Oriental magpie-robin, Long-tailed shrike, and White-breasted waterhen are resident birds, and Eurasian blackbird is winter migrant in Guangdong province. Individual insects, prawns, fish, and frogs were too small for contamination analyses, so multiple individuals were pooled into composite samples for each taxon in each sampling

campaign. Detailed information about each species is provided in Table S1 (“S” designates the tables in the Supplementary Material). All samples were transported to the laboratory in an ice box. After the species were identified, they were freeze-dried, homogenized in a stainless-steel blender, and stored at $-20\text{ }^{\circ}\text{C}$ until the time of analysis.

2.2. Chemical analysis

Details on the extraction, cleanup, and quantification of HOPs in insects and their predators were provided in our previous report (Liu et al., 2018a; 2018b). Briefly, following spiked with surrogate standards (CB 24, 82, and 198 for PCBs; BDE 118, BDE 128, 4-F-BDE 67, 3-F-BDE 153, and ^{13}C -BDE 209 for HFRs), a soxhlet extraction of 1 g of the lyophilized samples (whole or muscle) using hexane/dichloromethane (1/1, v/v) was conducted for 48 h. An aliquot of each extract (10%) was removed for gravimetric lipid determination and the remaining extract was purified using concentrated sulfuric acid and further cleansed in a multilayer Florisil silica gel column. The cleansed extracts were then concentrated to near dryness under a gentle nitrogen flow and finally reconstituted in isooctane (100 μL) for gas chromatograph mass spectrometer analysis (GC/MS). The recovery standards (PCB 30, 65, and 204; BDE 77, 181, and 205) were spiked prior to instrumental analysis. PCBs were analyzed by Agilent GC/MS using electron ionization in a selected ion monitoring mode; and PBDEs, DP, DBDPE, PBT, PBEB, HBB, and PBBs were analyzed using the electron capture negative ionization mode. Detailed procedures of instrumental analysis, quality assurance and quality controls are provided in the Supplementary Material.

2.3. Stable isotope analysis and trophic level determination

All samples for carbon isotope ($\delta^{13}\text{C}$) and nitrogen isotope ($\delta^{15}\text{N}$) analysis were freeze-dried and ground into powders. Approximately 0.5 mg of each sample was placed into a tin capsule and analyzed using a Flash EA 112 series elemental analyzer coupled with a Finnigan MAT ConFlo III isotope ratio mass spectrometer. Stable isotope abundances were calculated using the following equation:

$$X = (R_{\text{sample}}/R_{\text{standard}} - 1) \times 1000 \quad (1)$$

where X represents ^{13}C or ^{15}N , and $R_{\text{sample}}/R_{\text{standard}}$ is the $^{13}\text{C}/^{12}\text{C}$ or $^{15}\text{N}/^{14}\text{N}$ ratio of the sample. The precision for this technique is approximately $\pm 0.2\%$ (2 SD) for $\delta^{13}\text{C}$ and $\pm 0.5\%$ (2 SD) for $\delta^{15}\text{N}$.

The TLs of the biota species were calculated according to the following equation:

$$\text{TL}_{\text{consumer}} = (\delta^{15}\text{N}_{\text{consumer}} - \delta^{15}\text{N}_{\text{baseline}}) / \Delta\delta^{15}\text{N} + 2 \quad (2)$$

where $\text{TL}_{\text{consumer}}$ and $\delta^{15}\text{N}_{\text{consumer}}$ are the TL and stable nitrogen isotope abundance of the consumer, respectively. $\delta^{15}\text{N}_{\text{baseline}}$ is the stable nitrogen isotope abundance of the reference species: the beetle representing terrestrial species and the dragonfly larva representing aquatic species. $\Delta\delta^{15}\text{N}$ is the isotope enrichment factor and a common value of 3.4‰ was used (Post, 2002; Starrfelt et al., 2013).

2.4. Biomagnification and trophic magnification factors

BMF values were calculated based on a simple model that apporitions the contamination contributions from multiple prey species (Elliott et al., 2005; Chen et al., 2010). This model provides a more accurate estimation compared to simply calculating BMF values based on individual prey – predator food chains. The model uses the following equation:

$$\text{BMF} = C_{\text{predator}} / \sum_{i=1}^n (C_{\text{prey } i} \times f_{\text{prey } i}) \quad (3)$$

where C_{predator} is the geometric mean concentration of HOPs (ng g^{-1}

lipid weight) in predatory species; $f_{\text{prey } i}$ represents the contribution of prey type “i” to the predator’s total diet, and $C_{\text{prey } i}$ is the geometric mean concentration of HOPs (ng g^{-1} lipid weight) in the prey.

The values of TMF for aquatic and terrestrial food webs were estimated based on the linear regression analysis of logarithmically transformed lipid-normalized HOPs concentrations versus TL using Eq. (4). The values of TMF were calculated as the anti-ln of the slope (b) of the regression line based on the following equation:

$$\ln C_{\text{HOPs}} = a + b \times \text{TL} \quad (4)$$

$$\text{TMF} = e^b \quad (5)$$

where C_{HOPs} represents the HOPs concentrations, a is the y-intercept (constant), and b is the regression slope of $\ln C_{\text{HOPs}}$ versus TL. The statistical significance of the regression in Eq. (4) was defined as $p < 0.05$. A BMF or TMF value that is above one indicates that a chemical undergoes biomagnification, whereas a BMF or TMF value that is below one implies that a chemical is not accumulated by an organism or is metabolized (Fisk et al., 2001; Kelly et al., 2008).

2.5. Statistical analysis

Statistical analyses were performed using SPSS 19.0 and Origin 8.5. Simple linear correlation analysis was performed to investigate the relationship between HOPs concentrations and TLs in the terrestrial species. Multiple regression analysis (matrix analysis with the Kriging gridding method) was used to predict the relationships between TMFs (or BMFs) and the partition coefficients (K_{OW} and K_{OA}) of HOPs. Values of p below 0.05 were considered to be statistically significant.

3. Results and discussion

3.1. BMF in insect-dominated food chains

The dietary compositions of insectivorous amphibians, lizards, and birds are reported in previous studies based on field investigations (Wen, 1982; He, 1995; Zhu et al. 1999; Qiu et al., 2001) and details are provided in Table S2. Among the six orders of insects collected in this study, *Coleoptera* and *Lepidoptera* accounted for about 16% (Toad) to 65% (White-breasted waterhen) of the total diet of predators, while *Odonata* (0.27%) and *Mantodea* (0.40%) are only eaten by lizards. Other than the six orders of insects collected in the present study, other orders of insects, including *Diptera* (midges, flies, etc.), *Hymenoptera* (bees, ants, etc.) and *Homoptera* (aphids, psyllids, etc.) are also the food items of above predators and they accounted for 13–79% of the diets of predators (Table S2). Since these insects were not collected in this study, the geometric mean concentrations of HOPs in all collected insects were used to estimate the HOPs levels in uncollected insects, resulting in some uncertainty in the calculated BMF values.

Because fish, prawns, and watersnakes are not insect-eating predators and frogs (*Kaloula pulchra*) mainly feed on ants (which account for > 90% of their diet), the actual predator – prey relationships established in the present study were insects – birds, insects – toads, and insects – lizards. The concentrations of HOPs in the predators and most of the insects have been described in detail in the previous studies (Liu et al., 2018a, 2018b). Concentrations of HOPs in other insects including mantises, beetles, and crickets, as well as those published in previous studies were given in Table S3. Based on dietary composition, the BMF values of HOPs in insect-dominated food chains are listed in Tables 1 and S5.

The BMF values of HOPs in the four insectivorous birds range from 0.17 to 490 for insects – Oriental magpie-robins, 0.24 to 170 for insects – Eurasian blackbirds, 0.29 to 280 for insects – Long-tailed shrikes, and 2.1 to 76 for insects – White-breasted waterhen eggs, respectively. The BMFs for PCBs, PBDEs, PBBs, DP, and HBB in the four food chains are all greater than one (9.5–230), indicating significant biomagnification

from insects to insectivorous birds. However, DBDPE exhibit BMF values of less than one (0.17–0.43), implying biodilution. Some studies have highlighted the biomagnification of PCBs and PBDEs in insect – bird food chains, but little information regarding DP and alternative brominated flame retardants (BFRs) has been reported. Maul et al. (2006) found that the BMF values for PCBs range from 7.6 to 54 (19 for Σ PCBs) in the terrestrial insect – tree swallow nestling food chain. The insects in that study were considered to belong to one group and included *Coleoptera*, *Lepidoptera*, *Orthoptera*, *Hemiptera*, and *Hymenoptera*. Similar BMF values were reported for the terrestrial invertebrate (*Coleoptera*) – bird egg food chain (3.1–35 for PCBs) (Blankenship et al., 2005). Nie et al. (2015) reported that the BMF values for PBDEs in the insect – turtledove food chains ranged from 0.2 to 12, while those of the Σ PBDEs for the grasshopper – turtledove, butterfly – turtledove, dragonfly – turtledove, and ant – turtledove food chain were 1.6, 1.6, 2.2, and 3.6, respectively. The species of insects in the studies mentioned above were similar to those in our study, but the values of BMF for PCBs and PBDEs were obviously lower than those in the four insect – bird food chains considered in this study.

The BMF values for PCBs, PBDEs, PBBs, and DP range from 1.8 to 9.4 in the insect – toad food chain and 2.0 to 14 in the insect – lizard food chain (Table 1), suggesting biomagnification. However, the BMFs of PBT, PBEB, HBB, and DBDPE are less than one (0.21–0.75 for insects – toads and 0.20–0.45 for insects – lizards, respectively), implying biodilution. The BMF value for α -HBCD in insect – toad and insect – lizard food chains was also found to be lower than one (0.29 and 0.63, respectively) in our previous study (Liu et al., 2020). The BMFs for HOPs in insects – lizards are comparable to those in insects – toads, implying that the biomagnification potentials of these HOPs in the lizards were similar to those in the toads. Wu et al. (2009a, 2018) reported that the BMFs range from 0.1 to 40 for PBDEs and 1.8 to 2.7 for DP in the insect – frog (*Rana limnocharis*) food chain. These results are similar to those reported for the BMFs of PBDEs and DPs in the toads, but the observed BMF values for BDE 206, 207, 208, and 209 (2.3–40) are clearly greater than those calculated in this study (0.64–1.7).

The BMF values for HOPs (except for DBDPE) in toads and lizards were significantly lower than those in the four insectivorous birds ($p < 0.05$), which could be a result of species specificity. Birds are homeotherms, whereas toads and lizards are poikilotherms. Homeotherms generally have higher energy requirements and greater efficient metabolic capacity compared to poikilotherms (Hop et al., 2002; Hallanger et al., 2011; Walters et al., 2016). This differences in the BMFs of HOPs between poikilotherms and homeotherms have also

Table 1
Biomagnification factors (BMFs) for HOPs in the insect-dominated food chains from an e-waste recycling site in South China.

Congener	Toad	Lizard	Eurasian blackbird	Oriental magpie-robin	Long-tailed shrike	Waterbird egg
Σ PCBs ^a	9.4	2.9	30	81	98	38
Σ PBDEs ^b	1.8	2.0	15	59	45	17
Σ DP ^c	2.3	2.3	23	13	9.5	21
Σ PBBs ^d	9.3	14	31	140	230	15
PBT	0.30	0.45	1.5	1.7	0.59	na ^e
PBEB	0.21	0.20	2.0	0.82	1.0	4.2
HBB	0.75	0.32	9.6	8.9	4.9	34
DBDPE	0.19	0.34	0.24	0.17	0.43	na

^a Sum of 30 selected PCB congeners (CB18, 28/31, 49, 52, 74, 87/115, 95, 99, 101, 105, 110, 118, 128, 138, 146, 149, 153/132, 156, 163/164, 167, 170/190, 174, 180/193, 183, 187, 189, 194, 196/203, 206 and 209).

^b Sum of 15 PBDE congeners (BDE28, 47, 99, 100, 153, 154, 183, 196, 197, 202, 203, 206, 207, 208 and 209).

^c Sum of *syn*-DP and *anti*-DP.

^d Sum of PBB 153 and PBB 209.

^e Not available, because the compound was not detected in waterbird eggs.

Table 2
Trophic magnification factors (TMFs) and the *p* values for HOPs in aquatic and terrestrial food webs from an e-waste recycling site in South China.

Congener	Aquatic		Terrestrial		Congener	Aquatic		Terrestrial	
	TMF	<i>p</i>	TMF	<i>p</i>		TMF	<i>p</i>	TMF	<i>p</i>
PCB18	1.29	0.84	0.82	0.12	PCB209	9.71	< 0.01	3.30	< 0.01
PCB28/31	1.88	0.16	1.24	0.37	ΣPCBs ^a	5.74	< 0.01	4.04	< 0.01
PCB49	5.65	0.01	0.88	0.71					
PCB52	9.34	< 0.01	1.35	0.28	BDE28	0.79	0.81	3.27	< 0.01
PCB74	2.39	< 0.01	2.25	0.03	BDE47	4.09	0.01	8.78	< 0.01
PCB87/115	9.61	< 0.01	1.88	0.14	BDE100	7.22	< 0.01	7.24	< 0.01
PCB95	11.9	< 0.01	1.77	0.09	BDE99	1.40	0.36	9.22	< 0.01
PCB99	5.80	< 0.01	3.17	< 0.01	BDE154	8.33	< 0.01	6.29	< 0.01
PCB101	12.0	< 0.01	3.02	< 0.01	BDE153	2.41	0.02	9.35	< 0.01
PCB105	6.62	< 0.01	3.45	< 0.01	BDE183	0.98	0.99	5.45	< 0.01
PCB110	8.10	< 0.01	1.62	0.41	BDE202	3.22	0.03	2.65	< 0.01
PCB118	7.43	< 0.01	3.74	< 0.01	BDE197	1.10	0.88	1.69	< 0.01
PCB128	8.62	< 0.01	3.50	< 0.01	BDE203	0.65	0.44	3.97	< 0.01
PCB138	11.6	< 0.01	3.96	< 0.01	BDE196	0.94	0.93	3.69	< 0.01
PCB146	3.48	< 0.01	4.82	< 0.01	BDE208	0.30	0.02	1.21	0.53
PCB149	10.6	< 0.01	3.37	< 0.01	BDE207	0.33	0.07	1.10	0.78
PCB153/132	3.29	< 0.01	4.58	< 0.01	BDE206	0.27	0.02	1.66	0.16
PCB156	5.21	< 0.01	11.0	< 0.01	BDE209	0.34	0.08	1.63	0.07
PCB163/164	4.31	< 0.01	8.58	< 0.01	ΣPBDEs ^b	0.78	0.58	2.94	< 0.01
PCB167	4.22	< 0.01	3.95	< 0.01					
PCB170/190	4.39	< 0.01	7.75	< 0.01	<i>syn</i> -DP	0.43	0.04	1.63	< 0.01
PCB174/181	14.3	< 0.01	1.51	0.04	<i>anti</i> -DP	0.19	< 0.01	1.72	< 0.01
PCB180/193	5.18	< 0.01	8.24	< 0.01	ΣDP ^c	0.24	< 0.01	1.67	< 0.01
PCB183	4.76	< 0.01	6.00	< 0.01					
PCB187	3.86	< 0.01	4.54	< 0.01	PBB153	2.05	0.02	10.8	< 0.01
PCB189	3.07	0.01	3.75	< 0.01	PBT	0.20	0.71	0.91	0.75
PCB194	3.41	< 0.01	10.7	< 0.01	PBEB	0.77	0.52	1.36	0.36
PCB203/196	4.93	< 0.01	7.62	< 0.01	HBB	0.64	0.08	1.86	0.01
PCB206	6.66	< 0.01	8.00	< 0.01	DBDPE	0.23	< 0.01	1.20	0.51

^a Sum of 30 selected PCB congeners.

^b Sum of 15 PBDE congeners.

^c Sum of *syn*-DP and *anti*-DP.

been observed in the Barents Sea food chain (Hop et al., 2002), Arctic marine food chain (McKinney et al., 2012) and a coastal food chain (Haukås et al., 2010).

3.2. TMF in aquatic and terrestrial food web

According to the stable isotopes of nitrogen ($\delta^{15}\text{N}$) in Table S1, a wider range of $\delta^{15}\text{N}$ values were identified in three terrestrial birds (2.5–9.4‰). These values were even lower than those in the insects, indicating the variability in birds' diets in the present study. Meanwhile, the extremely small number of birds could also result in high uncertainty in the final data. Therefore, the terrestrial food web considered in this study is composed of terrestrial insects (beetles, grasshoppers, crickets, mole-crickets, butterflies, moths, mantises, and dragonflies) and lizards, but does not include terrestrial birds. The aquatic food web consists of dragonfly larva, prawn, fish, watersnake, and White-breasted waterhen egg. The trophic status of the aquatic and terrestrial species was elucidated utilizing the $\delta^{15}\text{N}$ and TL values of aquatic and terrestrial food web components were shown in Fig. S1. The calculated TMF values for HOPs based on the lipid-normalized concentrations in aquatic and terrestrial food webs are listed in Table 2.

The TMF values for PCBs in aquatic and terrestrial food webs range from 1.29 to 14.3 and 0.82 to 11.0, respectively (Table 2). The concentrations of ΣPCBs increase as TL increases in aquatic and terrestrial food webs (Fig. 1a, $p < 0.05$), and TMFs for ΣPCBs were 5.74 and 4.04, respectively. Some low-chlorinated PCB congeners (CB 18, 28/31, 49, 52, 87/115, 95 and 110) exhibit TMF values roughly equal to one and are not statistically significant ($p > 0.05$), indicating that these congeners were not biomagnified in present food webs (Table 2). Fremlin (2018) also found that most PCBs have TMF values that are greater than one (1.20–15.66) in a terrestrial food web (including insects and birds) in urbanized regions of Metro Vancouver, Canada. The

TMF values reported for PCB 28/31 (1.20) and PCB 110 (1.78) are very close to those identified in this study (1.24 for PCB 28/31 and 1.62 for PCB 110). The TMFs for PCBs in the freshwater and marine food webs were generally greater than one in previous studies (Wu et al., 2009b; McKinney et al., 2012; Yu et al., 2012), indicating that PCBs have trophic magnification potential in both aquatic and terrestrial ecosystems.

The TMF values for PBDE congeners in the terrestrial food web range from 1.10 to 9.35, with tri- to octa-BDEs (1.69 – 9.35) and ΣPBDEs (2.94) having TMF values that are significantly greater than one ($p < 0.05$) (Table 2 and Fig. 1b). For the aquatic food web, only BDE 47, 100, 153, 154, and 202 exhibited TMF values significantly greater than one ($p < 0.05$, Table 2). The TMF value of BDE 100 (7.22) is greater than that of BDE 99 (1.40), and the TMF value of BDE 154 (8.33) is also larger than that of BDE 153 (2.41) in the aquatic food web, which is consistent with the reported TMFs in previous studies (Wu et al., 2009b; Choo et al., 2019; Kobayashi et al., 2019). However, TMF of BDE99 (9.22) was higher than that of BDE100 (7.24) and TMF of BDE153 (9.35) was higher than that of BDE154 (6.29) in the terrestrial food web. These results demonstrated a difference in biomagnification potential of PBDE congeners between aquatic and terrestrial environments. This difference between the aquatic and terrestrial food web may be due primarily to the differences in the metabolism of PBDEs between aquatic (main fish) and terrestrial species. Previous studies demonstrated that PBDE congeners can debromination in most of fish species (Mizukawa et al., 2013; Luo et al., 2017). This will result in high bioaccumulation of PBDE congeners that are resistant to debromination or the products of debromination such as BDE 47, 100, and 154. However, The general oxidation metabolism of PBDEs in terrestrial organisms such as birds and mammals, including human beings, is more effective for lower brominated congeners than higher brominated congeners. Therefore, higher brominated PBDE

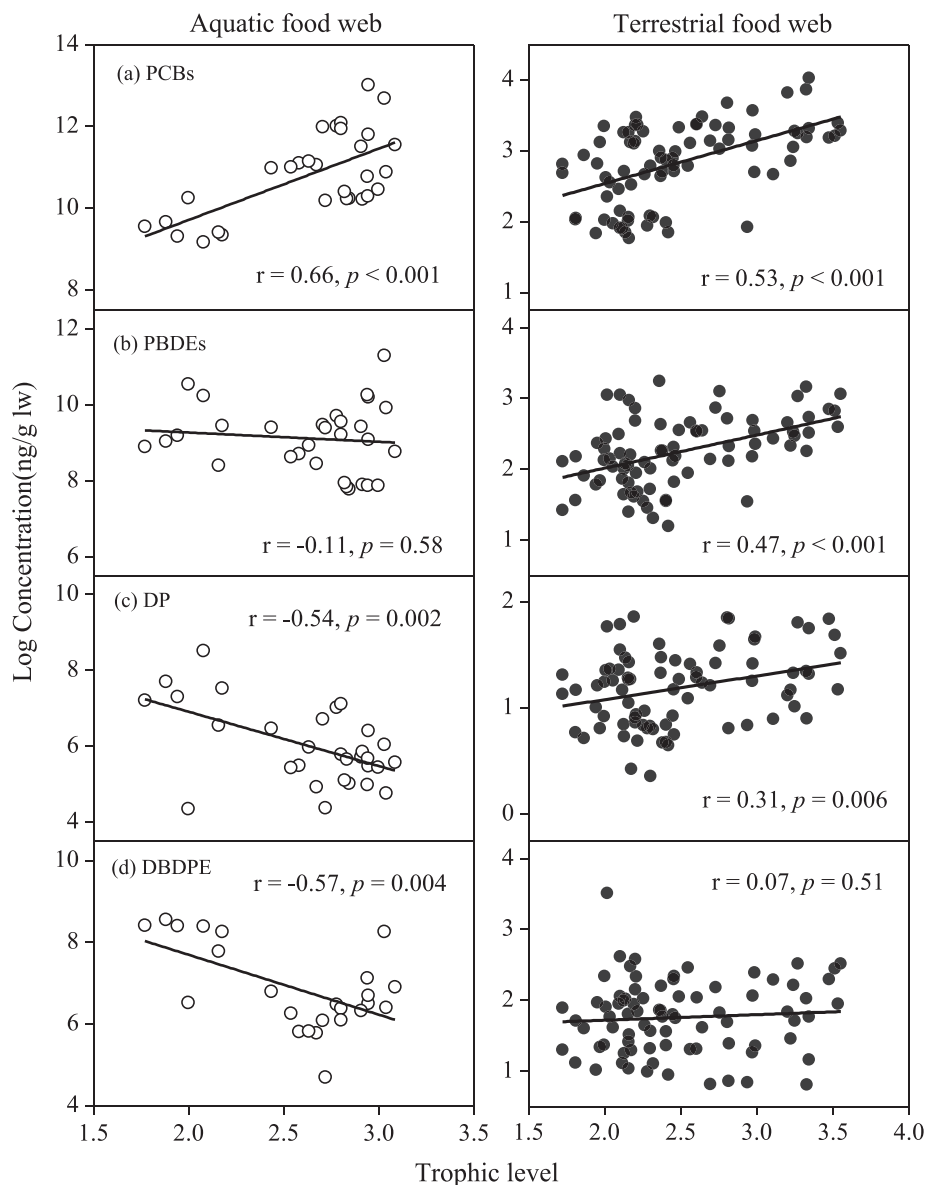


Fig. 1. Trophic transfer of PCBs (a), PBDEs (b), DP (c), and DBDPE (d) in aquatic and terrestrial food webs from an e-waste recycling site in South china.

congeners have greater biomagnification potential (Stapleton et al., 2009; Erratico et al., 2015). In this case, higher brominated PBDE congeners have greater biomagnification potential in terrestrial organisms, such as the predatory bird food chains (Voorspoels et al., 2007; Yu et al., 2011) and vegetation – caribou – wolf food chain (Morris et al., 2018). In this study, the terrestrial food web does not include birds, mammals, or other higher trophic level organisms, therefore significant biomagnification of lower brominated PBDE congeners can still be observed. Moreover, the dust may also play a role in the difference between aquatic and terrestrial food webs. Dust ingestion is considered to be an important pathway of terrestrial organisms exposure to PBDEs (Voorspoels et al., 2007). The concentrations of BDE47, 99, and 153 were higher than BDE100 and 154 in dust from the same e-waste site in previous study (Zheng et al., 2015).

In the terrestrial food web, the TMF values for DP, HBB, and PBB 153 (1.67, 1.86, and 10.8, respectively) are all statistically higher than one ($p < 0.05$, Table 2 and Fig. 1c), indicating trophic magnification. However, the TMFs for DP (0.24) and DBDPE (0.23) in the aquatic food web are statistically lower than one ($p < 0.05$, Fig. 1c and 1d), suggesting trophic dilution. Additionally, the TMF values for PBEB and PBT in aquatic and terrestrial food web were close to one, but not

statistically significant ($p > 0.05$, Table 2). To date, only a few studies have investigated the TMFs of such compounds in the aquatic food web. For example, Tomy et al. (2007) found that *anti*- and *syn*-DP were diluted with increasing TL in the Lake Ontario food web, and only *syn*-DP (TMF = 0.45, $p = 0.01$) showed trophic dilution in the Lake Winnipeg food web. However, Wu et al. (2009c) reported that DP was significantly biomagnified in a freshwater food web and that the TMF value of *syn*-DP (11.3) was greater than that of *anti*-DP (6.5). Sun et al. (2015) reported that the TMFs of DP and DBDPE in a mangrove food web were 2.31 and 0.85, respectively, but these values are not statistically significant ($p > 0.05$). Furthermore, a similar TMF value for DBDPE was found in the Lake Taihu food web (TMF = 0.37, $p = 0.012$) (Zheng et al., 2018), whereas significant trophic magnification was reported in the Lake Winnipeg food web (TMF = 8.9, $p < 0.05$) (Law et al., 2006). Regarding alternative BFRs, HBB was also found to be biomagnified (1.46), whereas PBT was biodiluted (0.61) in a freshwater food web. However, all these values are not statistically significant ($p > 0.05$) (Wu et al., 2010).

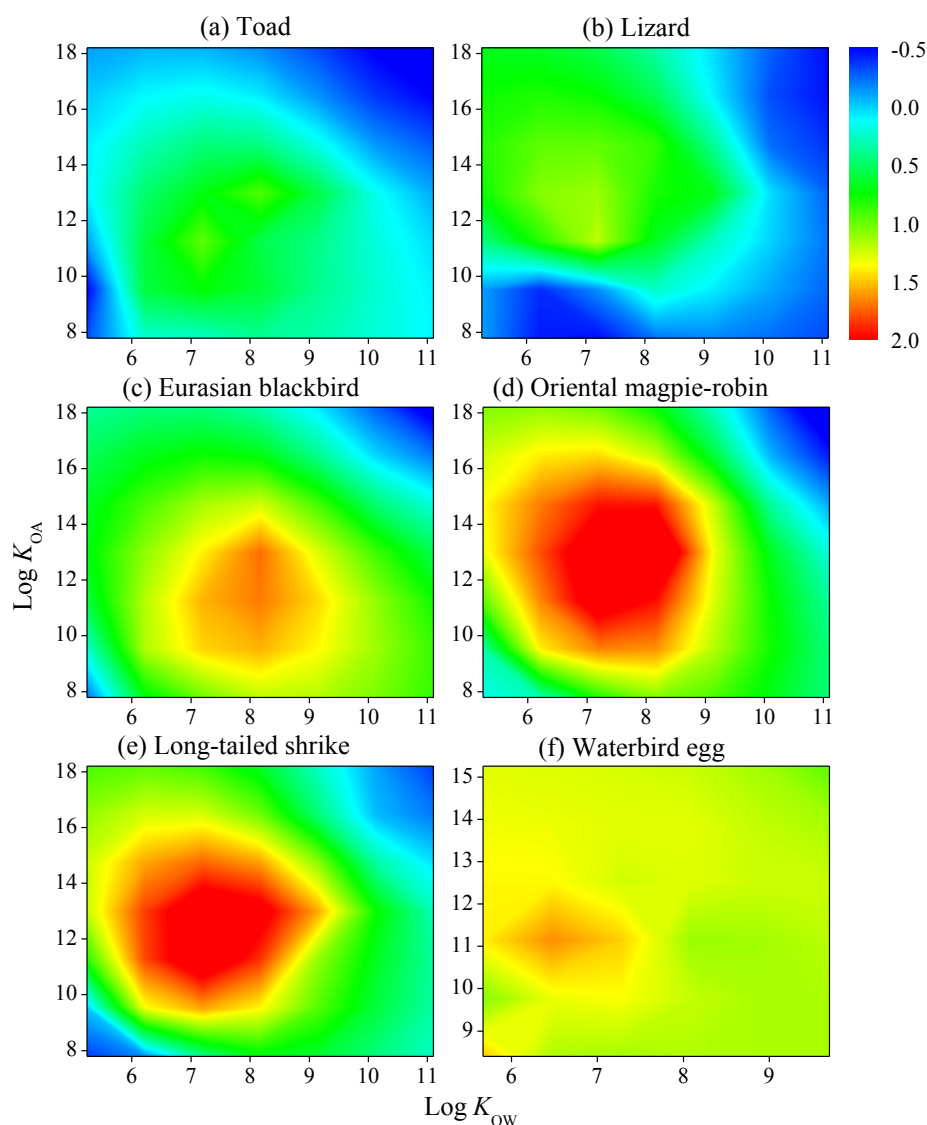


Fig. 2. Contour plots illustrating the relationship between $\log K_{OW}$ (x axis), $\log K_{OA}$ (y axis), with $\log BMFs$ (z dimension represented as colored contours) in the insect-dominated food chains. Red, yellow, and green indicate $BMFs > 1$ and blue indicate $BMFs < 1$. $\log K_{OW}$ values of HOPs were taken from Hawker and Connell (1998), Braekevelt et al. (2003), Sverko et al. (2011), and Covaci et al. (2011). $\log K_{OA}$ values were taken from Harner and Bidleman (1996), Wang et al. (2008), and Zhang et al. (1999; 2016). (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

3.3. Correlation between BMF/TMF and $\log K_{OW}/\log K_{OA}$

To acquire insight into the effects of chemical properties on the biomagnification behaviors of HOPs in different food chains, multiple regressions were performed to predict the relationships between $\log BMF$ and $\log K_{OW}$ and $\log K_{OA}$ of HOPs. The contour plots illustrate that both K_{OW} and K_{OA} can affect the biomagnification behaviors of chemicals in air-breathing organisms (Fig. 2). HOPs with $\log K_{OW} = 6-9$ and $\log K_{OA} = 10-16$ exhibit the greatest BMF values in the four insect-bird food chains, indicating a stronger biomagnification potential for the chemicals in insectivorous birds. Kelly et al. (2007) reported that organic contaminants with $\log K_{OW} > 5$ and $\log K_{OA} > 6$ have inherent biomagnification potential in the air-breathing organisms of the terrestrial, marine mammal and human food chains, which is consistent with our results. As mentioned above, the biomagnification potential of HOPs in toads and lizards was observed to be relatively lower than that in insectivorous birds, as shown in Fig. 2. Interestingly, K_{OW} and K_{OA} may have different mechanisms for the biomagnification behaviors of HOPs between toads and lizards. The adjusted R-square of the regression equation between $BMFs$ and $\log K_{OA}$ (0.53) is greater than that

between $BMFs$ and $\log K_{OW}$ (0.40) in lizards, but toads exhibit the opposite trend ($R^2 = 0.29$ for $\log K_{OA}$ and $R^2 = 0.41$ for $\log K_{OW}$, Fig. S2). This implies that the biomagnification behaviors in lizards are more heavily influenced by K_{OA} , and K_{OW} has a greater effect on toads. This may be due to the lizard being a terrestrial reptile and the toad being an amphibian.

The contour plots were also performed to predict the relationships between TMF and $\log K_{OW}$ and $\log K_{OA}$ of HOPs in aquatic and terrestrial food webs, as shown in Fig. 3. The results suggest distinct differences in the trophic magnification behaviors of HOPs between aquatic and terrestrial food webs. Similar to the insect-bird food chains, HOPs with $\log K_{OW} = 6-9$ and $\log K_{OA} = 10-16$ exhibit the greatest trophic magnification potential in the terrestrial food web. However, HOPs with $\log K_{OW} = 5-10$ and $\log K_{OA} < 12$ had relatively higher TMF values in the aquatic food web. The differences in biomagnification behaviors between aquatic and terrestrial food webs were also found for hexabromocyclododecane in some previous studies (Zhu et al., 2017; Liu et al., 2020). These results indicate that different bioaccumulation mechanisms of persistent halogenated organic pollutants between aquatic and terrestrial food webs. The elimination rate of

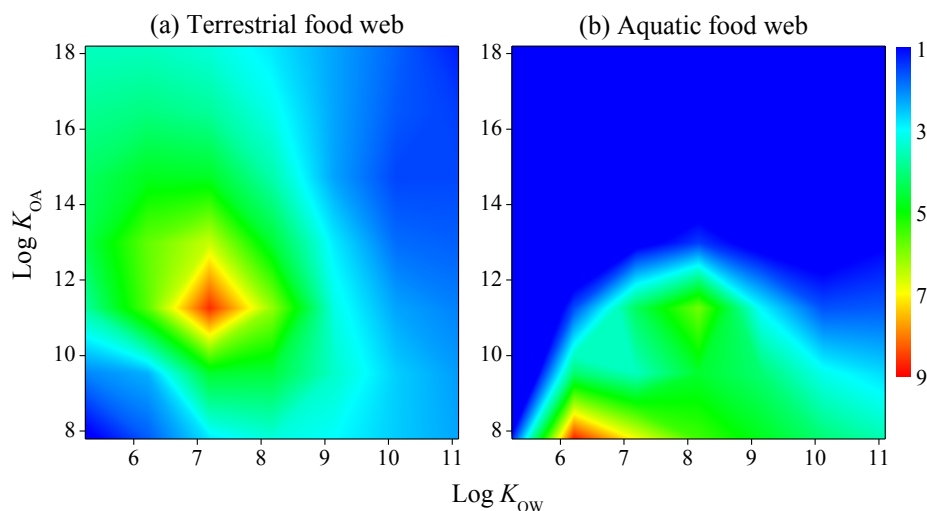


Fig. 3. Contour plots illustrating the relationship between $\log K_{OW}$ (x axis), $\log K_{OA}$ (y axis), with TMFs (z dimension represented as colored contours) in aquatic and terrestrial food webs.

chemicals which controlled by physical and chemical properties (K_{OW} and K_{OA}) may play an important role in the different bioaccumulation behaviors between aquatic and terrestrial ecosystems. Kelly et al. (2007) reported that elimination becomes sufficiently slow to cause biomagnification in water-respiring organisms if the $\log K_{OW}$ of the chemical exceeds 5, whereas the chemicals with a high $\log K_{OA}$ (> 6) could cause slow respiratory elimination in the air-breathing organisms. In addition, species-specific metabolism (such as fish and birds) and different exposure pathways (through water and sediment in aquatic organisms and via air, dust, and soil in terrestrial organisms) should also be considered (Kelly et al., 2007; Gobas et al., 2016; Zheng et al., 2016).

4. Conclusions

In this study, the BMF and TMF values of HOPs were calculated for insect-dominated food webs from an e-waste region in South China. The BMF values of HOPs (except DBDPE) in four insect – bird food chains were significantly higher than those in toads and lizards ($p < 0.05$), which may be due to the differences between poikilotherms and homeotherms. The TMF values for Σ PCBs and most congeners in aquatic and terrestrial food webs were greater than one, indicating that PCBs have trophic magnification potential in both aquatic and terrestrial ecosystems. The TMFs for Σ PBDEs and HBB in the terrestrial food web were all greater than one, but there was not biomagnification in the aquatic food web. Trophic dilution for DP and DBDPE was observed in the aquatic food web, whereas trophic magnification was found for DP in the terrestrial food web. Meanwhile, the contour plots of TMFs across combinations of $\log K_{OW}$ and $\log K_{OA}$ of HOPs were performed in aquatic and terrestrial food webs and illustrate distinct differences in the trophic magnification behaviors of HOPs between aquatic and terrestrial food webs. These results indicate that the bioaccumulation mechanisms of HOPs in aquatic organisms differ from those in terrestrial organisms, and further suggest that the bioaccumulation mechanism of contaminants in aquatic ecosystems may not be applicable to terrestrial ecosystems.

CRediT authorship contribution statement

Yu Liu: Data curation, Formal analysis, Writing - original draft. Xiaojun Luo: Conceptualization, Writing - review & editing. Yanghong Zeng: Methodology, Resources. Wenqing Tu: Software, Investigation. Mi Deng: Investigation. Yongming Wu: Visualization, Validation. Bixian Mai: Supervision, Funding acquisition.

Declaration of Competing Interest

The authors declared that they have no conflicts of interest to the work submitted.

Acknowledgments

This research was funded by the National Natural Science Foundation of China (Nos. 41673100, 41931290, and 41877386), Guangdong Foundation for Program of Science and Technology Research (Nos. 2017B030314057 and 2017BT01Z134), National Guided Local Science and Technology Development Project (No. 20192ZDD01001), and the Research and Development Project of Jiangxi Academy of Sciences (Nos. 2018XTPH121). This is contribution of IS-2837 from GIGCAS.

Appendix A. Supplementary material

Supplementary data to this article can be found online at <https://doi.org/10.1016/j.envint.2020.105674>.

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