Environmental Toxicology and Chemistry—Volume 41, Number 10—pp. 2395–2403, 2022
Received: 26 April 2022 | Revised: 30 June 2022 | Accepted: 9 July 2022

Environmental Chemistry

Bioamplification and Biomagnification of Polycyclic Aromatic Hydrocarbons and Halogenated Organic Pollutants in Moths from an Electronic Waste Recycling Site

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Abstract: Samples of *Asota caricae* (larvae, pupae, and adults) and its host plant leaf (*Ficus hispida*), larvae of *Aloa lactinea*, and mixed adult moths were collected from an electronic waste (e-waste) recycling site in south China. Polycyclic aromatic hydrocarbons, polychlorinated biphenyls, and polybrominated diphenyl ethers were found to exhibit concentrations of 420–1300, 100–860, and 7.6–49 ng/g dry weight, respectively. The different chemical compositions among the samples of the three groups indicated that at least two exposure scenarios occurred among the moths in the study area. A complex relationship between bioamplification factors and the octanol–water partition coefficient (log K_{OW}) was observed during metamorphosis, in which a positive relationship was evident for $5 < \log K_{OW} < 7$, whereas an inverse relationship occurred for log K_{OW} values outside of this range. The biomagnification factors (BMFs) calculated from the larvae of *Asota caricae* to the host plant were negatively correlated with log K_{OW} for all chemicals, differing completely from those obtained in previous studies. However, after metamorphosis, the correlation between BMF and log K_{OW} was found to coincide with that in previous studies. These results indicate that the biotransformation of pollutants in organisms played a key role in determining whether or not biomagnification occurred in organisms and highlight the potential application of the metabolic rate of chemicals in screening-level risk assessments of new chemicals. *Environ Toxicol Chem* 2022;41:2395–2403. © 2022 SETAC

Keywords: Bioamplification; Biomagnification; Moths; Metamorphosis; PAH; Halogenated organic pollutants

INTRODUCTION

Electronic waste (e-waste) is becoming a major environmental concern because of the harmful materials it contains and releases, including polychlorinated biphenyls (PCBs) and polybrominated diphenyl ethers (PBDEs), as well as the toxic chemicals generated during its disposal, such as polycyclic aromatic hydrocarbons (PAHs); PAHs, PCBs, and PBDEs are three classes of toxic organic pollutants that are frequently detected in soils and organisms in e-waste recycling sites (Leung et al., 2006; Wang et al., 2010; Wong et al., 2007). These hazardous chemicals are accumulated by organisms from environmental media and can cause serious environmental and health problems. Many studies have focused on the levels, distribution, and bioaccumulation of these pollutants in aquatic and terrestrial vertebrate organisms from e-waste recycling sites (Labunska et al., 2013; Liu et al., 2018b; Luo et al., 2007; Luo et al., 2009).

In the abiotic-to-biotic transfer process that persistent organic pollutants undergo, insects are important because of their large biomass and role as prey for predators. They can act as vectors for contaminant transfers of in-place pollutants to upper–food web components (Walters et al., 2008; Walters et al., 2010). However, biomagnification studies of contaminants have mainly been conducted on secondary (trophic level 3 to <4) and tertiary (trophic level 4 to <5) consumers, such as fish and mammals. Meanwhile, studies on insects are generally rare (Prince et al., 2020). The obtained results indicate that the octanol–water partition coefficient (log K_{OW}) is one of the key predictors of biomagnification (Arnot & Gobas, 2006; Kelly et al., 2007; Walters et al., 2011). Recently,

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This article includes online-only Supporting Information.
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DOI: 10.1002/etc.5432

Walters et al. (2016) proposed that the chemical solubility and metabolic transformation rate can be used to predict the biomagnification potential of organic pollutants in a more robust and accurate manner. The metamorphosis of insects provides an optimal window to gain insight into the effects of the biotransformation of chemicals on biomagnification potential because no input of chemicals or biotranformation is the sole factor in this process. In addition, few studies have investigated the bioaccumulation of contaminants from the plant to the larval stages of terrestrial invertebrates to date (Liu et al., 2020). Therefore, further research on lower trophic levels and on a broader range of taxa is required to develop a greater understanding of the key pathways through which pollutants are integrated into and accumulate in the food web.

Moths are a class of important terrestrial invertebrates that mainly feed on plant leaves. These metamorphotic insects play an important role in the trophic transfer of contaminants along the terrestrial food web. During metamorphosis, insects alter the chemical tracers used to denote food-web connections as well as toxic contaminants (Kraus et al., 2014). This concentration alteration in contaminants may be a key regulator of contaminant exposure and fluxes within the food web because insects can provide a predominant exposure pathway to consumers. There have been several studies regarding the alteration of contaminants during insect metamorphosis, and most have mainly focused on aquatic insects (Bartrons et al., 2007; Daley et al., 2011; Gewurtz et al., 2000; Huang et al., 2020; Reinhold et al., 1991; Viganò et al., 2007; Vives et al., 2005; Yu et al., 2013). Additional research is needed to demonstrate the effects of metamorphosis on the transport and transformation of chemicals in insects, especially in terrestrial insects. Moreover, no study has been conducted to investigate the effects of biotransformation on the biomagnification of chemicals in organisms during insect metamorphosis.

In the present study, moth samples and their host plant leaves in e-waste recycling sites were collected. The PAHs, PCBs, and PBDEs in the samples were subsequently determined. The present study aimed to (1) investigate the occurrence and accumulation of PAHs, PCBs, and PBDEs in moths; (2) assess the effects of metamorphosis on the transport from larvae to adult and biotransformation of chemicals; and (3) examine the biomagnification of target chemicals in plant–insect food chains and assess the effect of the biotransformation of chemicals on the biomagnification potential of chemicals.

MATERIALS AND METHODS

Sampling

The sampling sites were located in the woods around Longtang Town, Qingyuan County, Guangdong Province (23.6021°N, 113.0785°E) and have been intensely involved in e-waste processing for more than 10 years. The primitive recycling of e-waste has introduced toxic pollutants into the environment, resulting in high ecological risk for organisms in these regions (Guo et al., 2018; Wu et al., 2020). Insect and plant samples were collected between April and May of 2017.

With the use of tweezers, 38 larvae of *Asota caricae* were collected from the leaves of *Ficus hispida*. *Asota caricae* is a species of noctuoid moth in the family Erebidae, and *F. hispida* is one of the main host plants of *Asota caricae* (Wu & Gao, 2005). The day after the larvae of *Asota caricae* were captured, 18 larvae were found to transform into pupae. In a composite sample, four individual larvae and four to five individual pupae were pooled; in total, five pooled larvae and four pooled pupae samples were obtained. Adults of *Asota caricae* ($n = 30$, pooled to five composite samples) were also captured by sweeping nets in the woods, whereas 18 *Aloa lactinea* larvae were collected from the grass on the ground. Additional moths were captured using a black light trap installed on a tree. All moths collected via this method were mixed together, and three composite samples were obtained. The moths were not grouped into a single taxon because the sample size was too small for each species, and it was considerably difficult to identify and allocate each individual specimen to a specific species. From the trees in which the larvae of *Asota caricae* were collected, three *F. hispida* leaf samples were obtained. All samples were transported to the laboratory in an icebox. The samples were freeze-dried, homogenized using a stainless-steel blender, and stored at −20 °C until analysis.

Chemical analysis

The analytical procedures for the insect and leaf samples were the same as those used in previous studies (Huang et al., 2020). Leaves were rinsed with purified water to remove particulates on the leaf surface. Dry weights (0.2–0.8 g for insect samples and 1 g dry wt for leaf samples) were spiked with surrogate standards (chlorinated biphenyl [CB]24, CB82, and CB198 for PCBs; BDE118, BDE128, and ^{13}C-BDE209 for halogenated flame retardants; naphthalene-D_8, acenaphthylene-D_{10}, phenanthrene-D_{10}, and chrysene-D_{12} for PAHs). Then, the samples were Soxhlet-extracted, using a 200-ml Hex/Ace (1:1, v/v) for 48 h. The extract was evaporated to dryness and redissolved in *n*-hexane (10 ml). Then, one-tenth of the extract was used to determine the lipid content via gravimetric analysis. The remainder was subjected to gel permeation chromatography and eluted with dichloromethane/hexane (1:1, v/v) for lipid removal. Portions containing the target compounds from 90 to 280 ml were collected. The extract was then further cleaned on a multilayer Florisil silica gel column. The target chemicals were eluted with 60 ml of *n*-hexane/dichloromethane (1:1, v/v). After evaporation of the solvent to dryness, the extracts were redissolved in 200 µl of isooctane. The recovery standards (PCBs 30, 65, and 204; BDEs 77, 181, and 205; 2-fluorobiphenyl; *p*-terphenyl-d_{14}) were spiked prior to instrumental analysis.

The PCBs, including congeners chlorinated biphenyls 16, 18, 20/33, 22, 25/32, 26, 28/31, 40, 41, 42, 44, 45, 49, 52, 53, 63, 64, 66, 71, 74, 84, 85, 86/97, 87/115, 91, 92, 95, 99, 101, 105, 110, 118, 128, 135,138, 141, 146, 151, 153/132, 163, 164/175, 180/193, were determined from an Agilent 7890 gas chromatograph (GC) equipped with an Agilent 5975 mass spectrometer (MS), using electron ionization in selected ion monitoring (SIM) mode. Tri- to hepta-BDEs (BDEs 47, 100, 99, 153, 154, and 183) were analyzed using an Agilent 6890 GC coupled to an Agilent 5975 MS in electron capture negative ionization (ECNI) mode; BDE209 was analyzed using a Shimadzu model QP 2010 GC-MS (ECNI in SIM mode). Sixteen prior PAHs were analyzed using an Agilent 5890 GC coupled to an Agilent 5973 MS operating in SIM mode with electron ionization. The details of the instrumental analysis are provided in the Supporting Information.

Quality assurance and quality control

A procedural blank was run for each batch of 11 samples; PCB209, BDE209, naphthalene, and phenanthrene were detected in the procedural blanks. Their concentrations were <10% of those in samples with the exception of BDE209, for which levels in procedural blanks were up to 20% of those in samples. The recoveries of the surrogate standards (mean ± standard deviation) were 89 ± 8.1% for BDE118, 90 ± 10% for BDE128, 87 ± 10% for CB24, 86 ± 8.5% for CB82, 88 ± 10% for CB198, 24 ± 9.1% for naphthalene-D_8, 77 ± 5.3% for acenaphthylene-D_{10}, 86 ± 5.5% for phenanthrene-D_{10}, 96 ± 11% for chrysene-D_{12}, and 95 ± 11% for perylene-D_{12}. The final concentrations of the samples were corrected according to the procedural blanks. After subtracting the procedural blank, the concentrations of PCB209 in most of the samples were <0. Thus, PCB209 was undetectable in the samples. The recovery rates of naphthalene-D_8 were too low to be credible in the present study; thus, their associated data are not presented. The method detection limit (MDL) is defined as the average value plus three standard deviations for the target compounds detected in the procedural blanks. A signal-to-noise ratio of 10 was set as the MDL for analytes that could not be detected in blanks. The MDLs were between 15 and 104 pg/g dry weight for PCB congeners, between 3.4 and 320 pg/g dry weight for PBDE congeners, and between 50 and 500 pg/g dry weight for PAHs. The lipid contents were similar to each other (Table 1). Thus, the concentrations are expressed on a dry weight basis.

Data analysis

The bioamplification factors (BAmFs) were characterized by expressing the ratio of contaminant concentration in pupae to that in the larval stage, in adults to that in the pupae stage, and in adults to that in the larval stage of insects. The biomagnification factor (BMF) from plant leaves to larvae or adults of *Asota caricae* is calculated according to the formula $\mathrm{BMF} = C_{\mathrm{insect}}/C_{\mathrm{plant}}$, where C_{insect} is the concentration of chemicals in the larvae or adults of *Asota caricae* (nanograms per gram dry wt) and C_{plant} is the concentration of plant leaves (nanograms per gram dry wt).

Statistical analyses were performed using SPSS 20.0 and Origin 2018. One-way analysis of variance followed by Tukey's post hoc test was used to evaluate the differences in the levels of contaminants among the different insect species. Pearson's correlation analysis was conducted to investigate the correlation between BAmFs and log K_{OW}, and a simple linear regression analysis between BMFs and log K_{OW} was conducted. Statistical significance was set at $p < 0.05$.

RESULTS

Occurrence and composition of contaminants in moths

The statistical descriptions of the concentrations of PAHs, PCBs, and PBDEs in the insects and leaves of *F. hispida* are provided in Table 1 and Supporting Information, Table S1. No significant differences in PBDE concentrations were found among the different group samples (Table 1). The PAH levels in *Asota caricae* were significantly higher than those in *Aloa lactinea* ($p = 0.018$) and mixed moths ($p = 0.027$; Table 1), whereas PCB concentrations in the mixed moths were

TABLE 1: Concentrations of halogenated organic pollutants and polycyclic aromatic hydrocarbons in insects and leaf (nanograms per gram dry wt, mean ± standard deviation)

	Asota caricae			*Aloa lactinea*	Mixed moth	*Ficus hispida*
	Larvae	Pupae	Adult	Larvae	Adult	Leaf
Number[a]	5 (20)	4 (18)	5 (30)	3 (18)	3 (70)	3
Wet weight (g)[b]	2.78 ± 0.42	2.80 ± 0.55	1.29 ± 0.22	3.92 ± 0.33	0.79 ± 0.03	3.99 ± 0.32
Water content (%)	79 ± 1	76 ± 4	69 ± 6	84 ± 2	24 ± 0.0	74 ± 1
Lipid (%)[c]	12.2 ± 2.28	14.5 ± 3.73	17.0 ± 5.84	12.5 ± 1.24	16.6 ± 1.01	
PCBs[d]	187 ± 43	145 ± 7.3	186 ± 75	157 ± 2.3	722 ± 124	75 ± 1.6
PAHs[e]	647 ± 46	644 ± 119	940 ± 209	502 ± 83	575 ± 26	397 ± 45
PBDEs[f]	13.8 ± 3.4	11.4 ± 4.3	27.6 ± 15	17.2 ± 2.23	23 ± 4.1	150 ± 7.3

[a] Number in parentheses is the number of individual samples.

[b] Weight of composite sample.

[c] On the basis of dry weight.

[d] Sum of 42 PCB congeners (CB16, 18, 20/33, 22, 25/32, 26, 28/31, 40, 41, 42, 44, 45, 49, 52, 53, 63, 64, 66, 71, 74, 84, 85, 86/97, 87/115, 91, 92, 95, 99, 101, 105, 110, 118, 128, 135, 138, 141, 146, 151, 153/132, 163, 164/175, 180/193).

[e] Sum of 13 PAHs (acenaphthylene, acenaphthene, fluorine, phenanthrene, anthracene, fluoranthene, pyrene, benzo[*a*]anthracene, chrysene, benzo[*b*]fluoranthene, benzo[*k*]fluoranthene, benzo[*e*]pyrene, and benzo[*a*]pyrene).

[f] Sum of seven PBDE congeners (BDEs 47, 99, 100, 153, 154, 183, and 209).

PCB = polychlorinated biphenyl; PAH = polycyclic aromatic hydrocarbon; PBDE = polybrominated diphenyl ether.

significantly higher than those in the other sample groups ($p < 0.0001$).

The most abundant contaminants in *Asota caricae* and *Aloa lactinea* were PAHs, whereas PCBs were predominant in the mixed moths. The PAHs were dominated by phenanthrene in all samples, followed by fluorene, fluoranthene, and pyrene (Supporting Information, Figure S1). This pattern was highly similar to that found in organisms from Lake Redon (Pyrenees, Catalonia, Spain; Vives et al., 2005). The PCB congener profile can generally be classified into two types according to the dominant chlorinated biphenyl congener: CB18 was the most abundant congener in all samples of *Asota caricae*, whereas CB118 was predominant in the larvae of *Aloa lactinea* and mixed moths (Figure 1). The contributions of BDE209 to the total PBDEs in *Asota caricae* (72 ± 3.6% for larvae, 66 ± 4.5% for pupae, and 71 ± 8.2% for adults) were significantly higher than those in *Aloa lactinea* (36 ± 3.7%) and mixed moths (47 ± 5.2%). However, the contribution of BDE47 in the latter (~20%) two species was generally twice those (7.2 ± 2.2%, 10 ± 2.3%, and 8.3 ± 3.0% for larvae, pupae, and adults, respectively) found in *A. caricae* (Supporting Information, Figure S2).

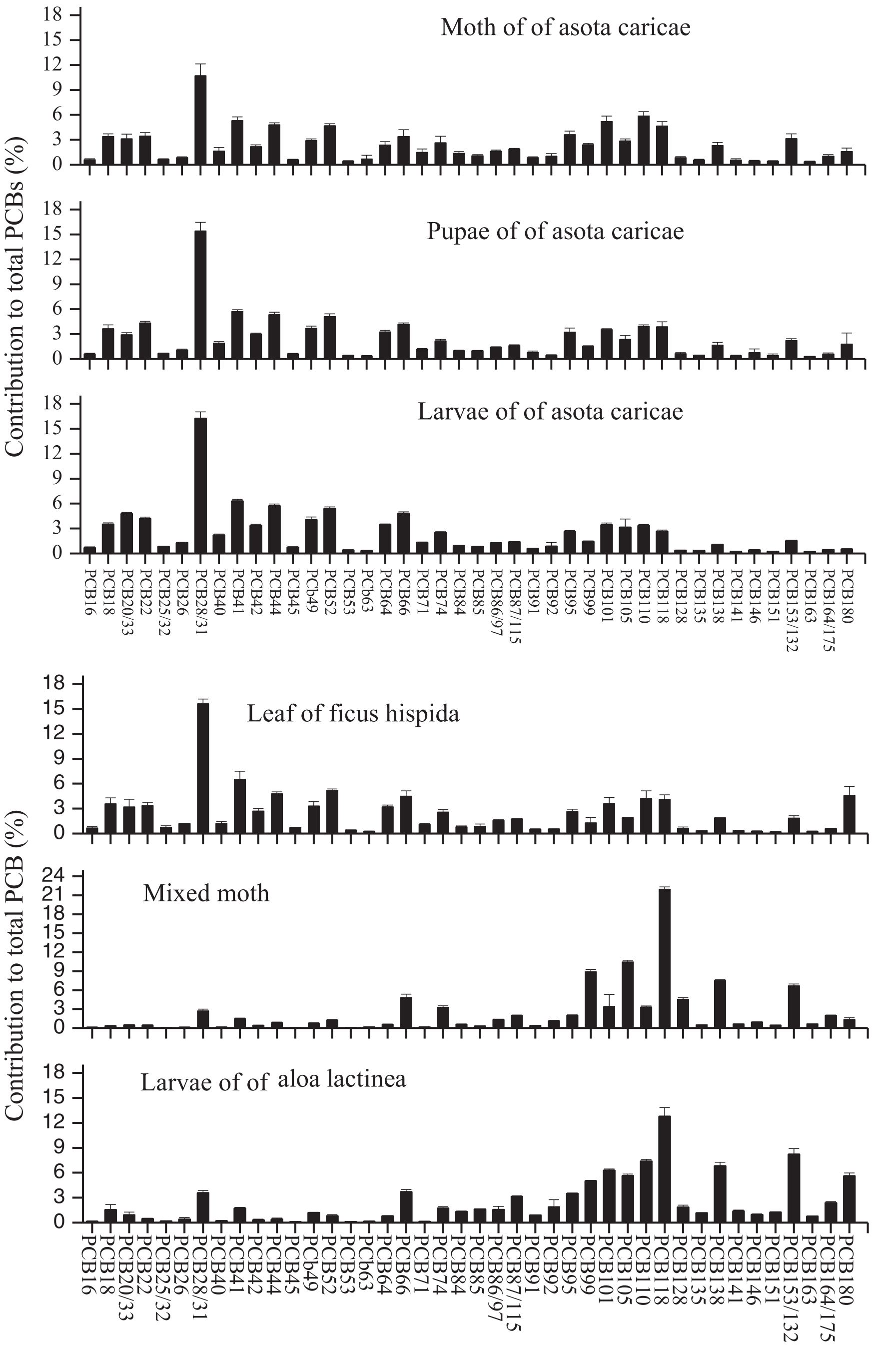

FIGURE 1: Polychlorinated biphenyl congener profiles in moth and plant leaf samples in the study area. PCB = polychlorinated biphenyl.

BAmFs of contaminants during metamorphosis in *Asota caricae*

During pupation (larvae to pupae), BAmF values varied from 0.61 (benzo[e]pyrene) to 1.34 (anthracene) for PAHs, from 0.48 (CB20) to 1.57 (CB146) for PCBs, and from 0.76 (BDE209) to 1.67 (BDE100) for PBDEs. During emergence (pupae to adults), the BAmFs of PAHs, PCBs, and PBDEs varied from 0.93 (anthracene) to 2.93 (benzo[e]pyrene), from 0.88 (CB28) to 2.60 (CB180), and from 1.29 (BDE100) to 2.71 (BDE209), respectively.

As for the pupation process, the BAmF of PAHs and PBDEs was negatively correlated with log K_{OW} but was not statistically significant ($p > 0.05$), whereas a significantly positive correlation was found for PCBs ($p < 0.0001$; Figure 2). Regarding the emergence process, a positive linear correlation between log K_{OW} and BAmF for log K_{OW} between 5 and 7 and a negative correlation for log K_{OW} outside of this range were observed (Figure 3). During the entire metamorphosis process, the BAmF (C_{adult}/C_{larvae}) showed a nonlinear correlation with the log K_{OW} of chemicals, which was closer to the correlation observed during the emergence process (Supporting Information, Figure S3). This indicates that alternation occurring in the emergence process plays a more important role for BAmFs than the pupation process for the entire metamorphosis.

BMFs of pollutants in *Asota caricae*

To gain insight into the effects of metamorphosis on the biomagnification of pollutants in insects, two sets of BMF data were calculated. The BMFs of chemicals from larvae of *Asota caricae* to *F. hispida* leaves were first calculated to investigate the transfer of chemicals from plants to insects. The BMF values for PAHs (from 1.30 for benzo[*a*]pyrene to 4.02 for acenaphthylene) and PCB congeners (from 1.30 for CB180 to 4.33 for CB40) were all >1, indicating biomagnification. The BMF values for PBDE congeners varied from 0.07 (BDE209) to 2.1 (BDE183). A negative correlation between BMF and log K_{OW} was found for all chemicals (marginal significance for PAHs, $p = 0.09$; significance for PCBs, $p < 0.001$; Figure 4). When we used the concentrations of chemicals in the adults of *Asota caricae* to recalculate the BMF, a significant linear positive correlation between BMF and log K_{OW} for PCBs was observed (Supporting Information, Figure S4), which was completely antithetical to that derived from the data on larvae of *Asota caricae*. Evidently, the alternation of contaminants occurring during metamorphosis was responsible for the observed reversal.

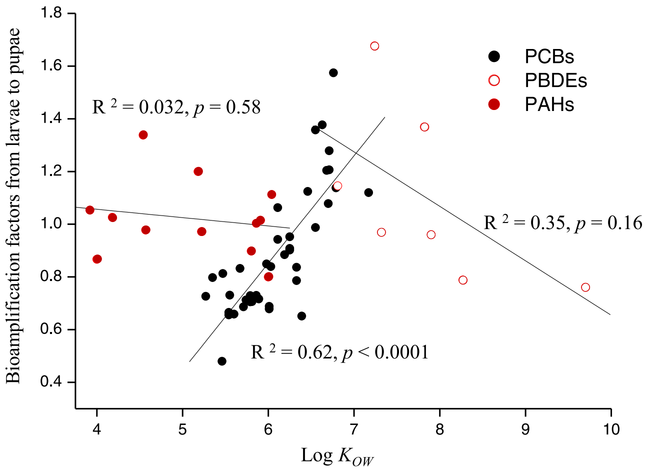

FIGURE 2: Relationship between bioamplification factors and log octanol–water partition coefficient for polycyclic aromatic hydrocarbons, polychlorinated biphenyls, and polybrominated diphenyl ethers from larvae to pupae of *Asota caricae*. PCBs = polychlorinated biphenyls; PBDEs = polybrominated diphenyl ethers; PAHs = polycyclic aromatic hydrocarbons; K_{OW} = octanol–water partition coefficient.

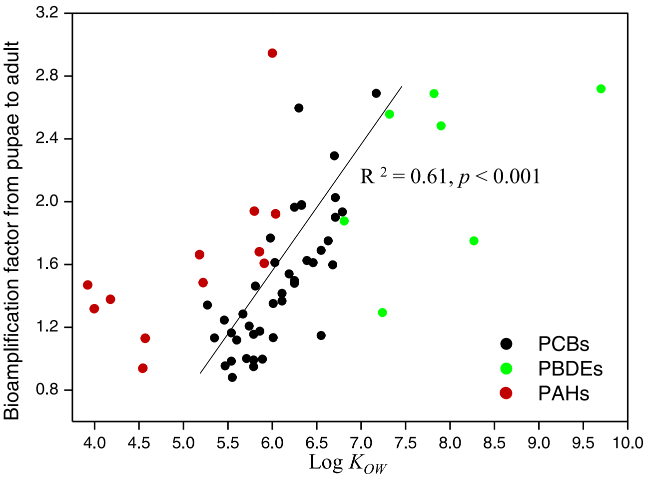

FIGURE 3: Relationship between bioamplification factors and log octanol–water partition coefficient for polycyclic aromatic hydrocarbons, polychlorinated biphenyls, and polybrominated diphenyl ethers from pupae to adult of *Asota caricae*. PCBs = polychlorinated biphenyls; PBDEs = polybrominated diphenyl ethers; PAHs = polycyclic aromatic hydrocarbons; K_{OW} = octanol–water partition coefficient.

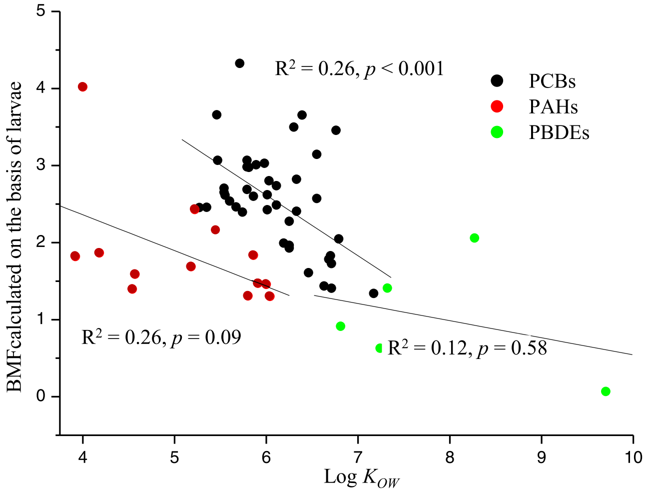

FIGURE 4: Correlation between biomagnification factors and log octanol–water partition coefficient for polycyclic aromatic hydrocarbons, polychlorinated biphenyls, and polybrominated diphenyl ethers in the plant leaf–*Asota caricae* food chain. PCBs = polychlorinated biphenyls; PBDEs = polybrominated diphenyl ethers; PAHs = polycyclic aromatic hydrocarbons; K_{OW} = octanol–water partition coefficient.

DISCUSSION

Different contaminant compositions revealed at least two different exposure scenarios for moths in the study area

A taxon-specific contaminant composition was found in the present study, indicating different exposure scenarios among lepidoptera in the study area. Differences in feeding preferences, metabolic capacities, and pollution sources could lead to the taxon-specific contaminant composition (Heintzman et al., 2015; Walters et al., 2018). In the present study, variable diet very likely caused the variability in pollutant patterns among the different species of insects.

In addition to collecting *F. hispida* leaf samples (host plant of the larvae of *Asota caricae*) in the present study, grass, water, and soil samples and another host plant for moth larvae (*Psidium guajave* leaf) were collected from the study area in our previous study (Liu et al., 2018a). The congener profiles of PCBs and PBDEs in the grass, similar to that in the *F. hispida* leaf in the present study, were dominated by CB28 and BDE209 (97%), respectively. However, chlorinated biphenyls 110, 118, 138, 153, and 28 dominated in the *P. guajave* leaf and soil samples (Supporting Information, Figure S5), and the contribution of BDE47 (6%) in the *P. guajave* leaf was approximately 10 times that of grass (0.6%) and the *F. hispida* leaf (0.7%; Supporting Information, Figure S6). Thus, there were at least two different pollution compositions in the host plants of moth larvae in the study area. One composition was represented by grass and the *F. hispida* leaf and was characterized by a high abundance of CB18 and BDE209. The other was represented by the *P. guajave* leaf, which was characterized by relatively high abundances of CB118 and BDE47. This was the main cause for the taxon-specific contaminant composition observed in the present study. The variable contaminant compositions in different host plants may have contributed to the diverse deposition mechanisms of pollutants in the plants (McLachlan, 1999; Tian et al., 2012) and the high heterogeneity of the environment pollution in the e-waste dumping sites.

BAmFs of contaminants during metamorphosis in Asota caricae

The BAmF of pollutant during metamorphosis was the combined result of an increase in concentration from the loss of body weight and a decrease in concentration on account of the metabolism. The average dry weights of the larvae, pupae, and adults of *Asota caricae* were 0.139, 0.100, and 0.07 g, respectively. Ideally, the expected BAmF would be 1.39 and 1.43 in the pupation and emergence processes, respectively, if no metabolism occurred. However, the measured BAmFs were less than the expected BAmF for most of the chemicals during the pupation process.

The biotransformation of contaminants occurring during pupation was responsible for the low BAmF values. Vives et al. (2005) reported a significant decrease when comparing the PAH concentrations of Chironomidae between the pupal and larval stages, with a BAmF of 0.24 between these stages. The BAmF was less than that in the present study (1.00), implying a stronger metabolism of PAHs in Chironomidae than in *Asota caricae*. A smaller measured BAmF than expected was also reported in our previous laboratory experiment, which exposed silkworms to PCBs and PBDEs (Huang et al., 2020).

Significant positive correlations between BAmFs and log K_{OW} were found for PCBs in both pupation and emergence processes (Figures 2 and 3). Similar positive correlations were also found in the silkworms exposed to PCBs in the laboratory experiment (Huang et al., 2020) and in Limnephilidae in a field monitoring study (Supporting Information, Figure S7; Bartrons et al., 2007). However, conflicting results were also reported. For example, Daley et al. (2011) found no significant correlations between BAmFs of PCBs and log K_{OW} for mayflies from the subimago to the imago state. In our laboratory exposure experiment, a positive linear correlation was found in the high-dose exposure group but not in the low-dose exposure group during the silkworm pupation process. Furthermore, a positive linear correlation was observed in female silkworms but a negative correlation in males during the emergence process (Huang et al., 2020). Negative correlations between BAmFs of PCBs and log K_{OW} were also found in Ceratopogonidae (Supporting Information, Figure S7; Bartrons et al., 2007). All of the above results indicate that the correlations between BAmF and log K_{OW} appear to be specific to chemical and taxonomical compositions and are concentration-dependent to an extent. Sex could be also a confounding factor for the observed correlations.

Liu et al. (2018a,b) observed a common alteration in the pollutant pattern (mainly PCBs and halogenated flame retardants) in different insects (dragonfly, butterflies and moths, grasshoppers, and litchi stinkbugs) during metamorphosis (adult to larvae). Kraus et al. (2014) reviewed the literature on studies of alternation of contaminants and chemical tracers during insect metamorphosis and found that the BAmF first decreased with log K_{OW} at log $K_{OW} < 5$, then increased with log K_{OW} when $5 < \log K_{OW} < 7$; finally, it decreased again with increasing log K_{OW}. The correlation between BAmF and log K_{OW} in the present study was consistent with the prediction of Kraus et al. (2014) and similar to that observed by Liu et al. (2018a) to a certain extent (Supporting Information, Figure S3). These results imply that there is a common mechanism for regulating the alternation of contaminants during insect metamorphosis.

Although the actual reasons for the inconsistent results observed among different studies are not currently known, the occurrence of biotransformation during metamorphosis could be a key factor in regulating the alternation of contaminants. During moth metamorphosis, no feeding behavior occurred. The inputs of contaminants are negligible. Generally, PCB metabolism in organisms decreased with increasing chlorinated degree. Thus, a positive correlation between BAmF and log K_{OW} for PCBs was observed. It is argued that adult moths can obtain contaminants by sucking the sap from the flowers and shoots of the host plant. Our previous study revealed that insects that suck the sap generally exhibited a much higher abundance of low log K_{OW} PCB congeners than those that feed on the plant leaf (Liu et al., 2018a), which is contrary to the decreased abundance of low–log K_{OW} PCB congeners evident in the present study. The decreased BAmF for BDE209 could be caused by elimination via exuviate because relatively high concentrations of BDE209 were found in the exuviate of silkworms in laboratory exposure experiments (Huang et al., 2020). Thus, both elimination by exuviate and biotransformation shaped the alternation of chemicals during metamorphosis.

Bioamplification reverses the correlation between BMF and log K_{OW} for PCBs

The BMF values calculated on the basis of the larvae of *Asota caricae* were negatively correlated with the log K_{OW} in the present study. High BMF values were found in chemicals with low log K_{OW}, which completely differed from the results of previous studies (Arnot & Gobas, 2006; Kelly et al., 2007; Walters et al., 2011). Most of the previous studies revealed that organic compounds with log K_{OW} <5 have a lower potential for biomagnification and that chemicals with $5 < \log K_{OW} < 7$ have the highest potential (Verhaert et al., 2017; Walters et al., 2016).

In a model developed by Thomann and Komlos (1999), biotransformation and decreased transfer efficiency from the gut to the organisms were two important factors in the presence of a low bioaccumulation factor. Biotransformation cannot be used to explain the diminished BMF for chemicals with a high log K_{OW} in the present study because PCBs with a low log K_{OW} showed a higher biotransformation rate than PCBs with a high log K_{OW}, as demonstrated during metamorphosis. Thus, low gut uptake efficiency should be the reason for the decrease in BMF with increasing log K_{OW}. Our previous laboratory experiment using silkworms provided direct evidence to support this hypothesis. The uptake efficiency of PCBs and brominated flame retardants linearly decreased with increasing log K_{OW} (Huang et al., 2020). Inverse relationships between the uptake efficiency and log K_{OW} have also been reported in other animals, such as fish (Zeng et al., 2012), doves (Drouillard & Norstrom, 2000), and human beings (Moser & McLachlan, 2001). This may be common when animals absorb pollutants from the gut to the organism. Thus, the observed inverse relationships between BMF and log K_{OW} based on the larvae of *Asota caricae* reflect the gut uptake efficiencies of chemicals. Elimination via biotransformation plays a minor role in this stage, possibly because of the poor biotransformation of pollutants in the larvae of *Asota caricae*, just as with the higher BAmF of PAH in *Asota caricae* than in Chironomidae (Vives et al., 2005).

When adult *Asota caricae* were used to recalculate the BMF, the correlation between BMF and log K_{OW} was found to coincide with those obtained from modeling studies, laboratory experiments, and field studies. It goes without saying that changes in pollutants during metamorphosis, such as elimination via biotransformation for PCBs, were responsible for the reversion, as mentioned previously.

The K_{OW} value is proposed as one of the key predictors of biomagnification. This could be applicable for bioconcentration processes, such as what occurs in the partition between water and organisms. However, it does not seem to be a suitable predictor for the biomagnification process. The biotransformation that occurred in the organisms may be more important than log K_{OW} in determining whether or not chemical biomagnification takes place in organisms. The observed significant positive correlations between BMF and log K_{OW} for PCBs in previous studies could be only a coincidence, owing to the metabolism potential of PCB congeners being positively correlated with log K_{OW}. Of course, the above statement requires more evidence because our understanding on the biotransformation of pollutants in insects is very scarce.

CONCLUSION

In the present study, moth and its host plant samples were collected from an e-waste-contaminated area and the PAHs, PCBs, and PBDEs in these samples measured. The contaminant pattern observed among moth samples implied that different exposure scenarios exist even in a small region. The observed correlation between BAmF during metamorphosis and log K_{OW} was similar to those in previous studies. Values of BMF calculated from larvae of moth to host plant negatively correlated with log K_{OW}, which was completely different from the results of previous studies. On the other hand, the correlation between BMF calculated from adult moths to host plant and log K_{OW} was consistent with those in previous studies. This result indicated that biotransformation during metamorphosis plays a crucial role in characterizing the correlation between the BMF and log K_{OW} in terrestrial moths, highlighting the importance of biotransformation in determining the bioaccumulation potential of chemicals.

Supporting Information—The Supporting information are available on the Wiley Online Library at https://doi.org/10.1002/etc.5432.

Acknowledgment—The present study was financially supported by the strategic Priority Research Program of the Chinese Academy of Science (XDPB2001), the National Nature Science Foundation of China (41877386, 41931290), the Local Innovative and Research Teams Project of the Guangdong Pearl River Talents Program (2017BT01Z134), and the Guangdong Foundation for the Program of Science and Technology Research (2020B1212060053, 2019B121205006). This is a contribution No IS-3225 and SKLOGA202004 from GIGCAS.

Disclaimer—The authors declare no conflict of interest.

Author Contributions Statement—**Luo Xiao-Jun**: Conceptualization; Data analysis; Writing—original draft. **Li-Qian Huang**:

Investigation; Writing—review & editing. **Ke-Lan Guan**: Investigation; Writing—review & editing. **Xue-Meng Qi**: Investigation; Writing—review & editing. **Yan-Hong Zeng**: Writing—review & editing; Project administration. **Bi-Xian Mai**: Resources; Supervision.

Data Availability Statement—Data, associated metadata, and calculation tools are available in the Supporting Information.

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