

Aquatic bioaccumulation and trophic transfer of tetrabromobisphenol-A flame retardant introduced from a typical e-waste recycling site

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Abstract While the flame retardant chemical, tetrabromobisphenol-A (TBBP-A), has been frequently detected in the environment, knowledge regarding its species-specific bioaccumulation and trophic transfer is limited, especially in the highly contaminated sites. In this study, the components of an aquatic food web, including two invertebrates, two prey fish, and one predator fish, collected from a natural pond at an electronic waste (e-waste) recycling site in South China were analyzed for TBBP-A, using liquid chromatography-tandem mass spectrometry. The aquatic species had TBBP-A concentrations ranging from 350 to 1970 pg/g wet weight, with higher concentrations in the invertebrates relative to the fish species. Field-determined bioaccumulation factors of TBBP-A in the two aquatic invertebrates were nearly or greater than 5000, suggesting that TBBP-A is highly bioaccumulative in the two species. The lipid-normalized concentrations of TBBP-A in the aquatic species were negatively correlated with the trophic levels determined from stable nitrogen isotope ($\delta^{15}\text{N}$) ($r=-0.82$, $p=0.09$), indicating that this compound experienced trophic dilution in the current food web.

Keywords Tetrabromobisphenol-A (TBBP-A) · Brominated flame retardants · Aquatic food web · Trophic transfer · Electronic waste

Introduction

Brominated flame retardant (BFR) chemicals are extensively used in a diverse array of products, including textiles, plastics, and electrical and electronic equipment, to inhibit or impede flammability. Tetrabromobisphenol-A (TBBP-A) was reported as one of the most used BFRs in the world, covering approximately 60 % of the global BFR usage (Covaci et al. 2009; Liu et al. 2016). The European BFR Industry Panel reported the worldwide TBBP-A production to be 170,000 tons in 2004 and stated that this value is increasing (EBFRIP 2007). Due to its large use and environmental persistence, TBBP-A is an ubiquitous environmental contaminant that is detected in both abiotic and biotic matrices (Covaci et al. 2009; Liu et al. 2016). TBBP-A is primarily used as a reactive flame retardant in epoxy, polycarbonate, and phenolic resins used in printed circuit boards but also has additive applications in several types of polymers such as acrylonitrile butadiene styrene (ABS) resins that are mainly used in automotive parts, pipes and fittings, and electrical and electronic equipment (Covaci et al. 2009). With the restriction on OctaBDE mixture, a polybrominated diphenyl ether (PBDE) flame retardant (UNEP 2010), TBBP-A is considered an alternative additive BFR to OctaBDE in ABS (Covaci et al. 2009). TBBP-A concentration in this application varies between 10 and 20 % (by weight) (Covaci et al. 2009). Currently, there are no restrictions on the production of TBBP-A. Hence, the additive applications of TBBP-A as an alternative to the discontinued BFRs (e.g., OctaBDE mixture) could increase in the future. As an additive BFR, it can be

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released to the environment from the polymeric products during the use of TBBP-A-containing products and recycling of these products, resulting in an exposure risk in human and wildlife (Covaci et al. 2009). Although information pertinent to the undesirable effects of TBBP-A is limited, studies have demonstrated that TBBP-A can act as an endocrine disruptor (Fini et al. 2007; Guyot et al. 2014; Grasselli et al. 2014).

TBBP-A has been subject to risk assessments of its use within the European Union. The Human Health Risk Assessment Report concluded that there was no risk to human health in relation to its primary reactive uses, such as in the epoxy resins used in printed circuit boards (European Chemicals Bureau 2006). However, the Environmental Risk Assessment Report for TBBP-A expressed concern in some scenarios for water and terrestrial compartments when TBBP-A is used as an additive BFR (European Chemicals Bureau 2008). The risk assessment by the European Union did not assess the worst-case scenarios such as e-waste recycling sites, primarily in Asia and Africa, with potentially higher levels of exposure to both humans and wildlife (Law 2009). Knowledge regarding the environmental fate and effects of TBBP-A in these worst-case scenarios is rather obscure.

Elevated levels of BFRs including TBBP-A have been reported in the environment and wildlife from e-waste recycling sites in Asia (Huang et al. 2014; Matsukami et al. 2015; Wang et al. 2015; He et al. 2013). More recently, Tang et al. (2015) reported high TBBP-A concentrations in a prey fish and a predator fish from an e-waste recycling site in South China. Interestingly, the authors found that the predator fish contained lower concentrations of TBBP-A than the prey fish, indicating a relatively low concentration in organisms at high trophic level (TL). This is contrary to those found for other BFRs (e.g., PBDEs) for which higher concentrations were generally detected in organisms at high TL due to the biomagnification of these chemicals (Wu et al. 2009). The observation suggests that TBBP-A may experience a distinct bioaccumulation behavior from other BFRs. However, the trophodynamics of TBBP-A in a whole aquatic food web, one of vital criteria for assessing the ecological risk of the chemical, has not been investigated.

The primary objective of this study was to explore the trophic transfer of TBBP-A in an aquatic food web from an e-waste recycling site in South China, one of the “worst-case scenarios” of TBBP-A. Additionally, TBBP-A levels were also determined in the dissolved phase of water. Field-determined bioaccumulation factors (BAFs) of TBBP-A were estimated in the aquatic species, to assess the species-specific bioaccumulation of this compound. This study represents the first example where the food-web transfer of TBBP-A is studied. This information will improve our understanding of the aquatic bioaccumulation and trophic transfer of TBBP-A in the worst-case scenarios.

Methods and materials

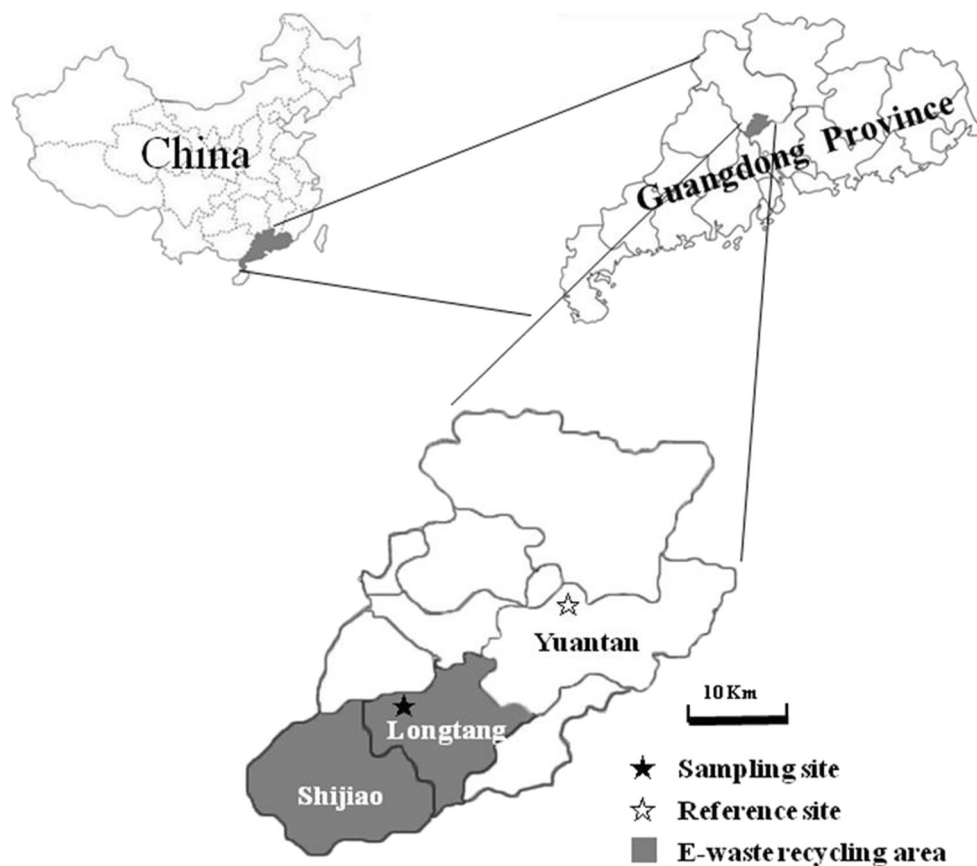
Sampling

Full details of the water and aquatic species sampling have been previously described (Wu et al. 2008). In brief, water and aquatic organisms representing different trophic levels of the food web were concurrently collected from a natural pond at an e-waste recycling site in South China (Fig. 1) in 2007. Reference samples (mud carp, *Cirrhinus molitorella*; $n=5$) were also collected from another pond approximately 20 km away from the e-waste recycling site (Fig. 1), representative TBBP-A level in fish without e-waste pollution. The aquatic species examined at the e-waste recycling site included two invertebrate species, i.e., the Chinese mystery snail (*Cipangopaludina chinensis*, $n=43$) and the prawn (*Macrobrachium nipponense*, $n=7$), two prey fish species, i.e., mud carp (*C. molitorella*, $n=12$) and crucian carp (*Carassius auratus*, $n=18$), and one predator fish species, northern snakehead (*Ophicephalus argus*, $n=6$). The Chinese mystery snail is one of the diet items of crucian carp (Yie et al. 1992), while prawn, mud carp and crucian carp are the main prey species of northern snakehead (Tian 1997). Some samples (e.g., Chinese mystery snail, prawn, and mud carp) were too small for individual contaminant analyses and were therefore pooled. The biota samples were whole body homogenized and stored in $-20\text{ }^{\circ}\text{C}$ until further analysis.

Sample extraction and cleanup

Biota samples were extracted according to the method described by Tang et al. (2015), with minor modifications. Briefly, samples were spiked with internal standard (^{13}C -TBBP-A) and ground with ashed anhydrous sodium sulfate in a glass mortar. The mixture was transferred to a glass extraction thimble and Soxhlet extracted with hexane/acetone (1/1, v/v) for 48 h. An aliquot of the extract was used to lipid content determination by gravimetric method. The remainder was subjected to gel permeation chromatography (GPC) to remove lipids and other interfering compounds. The extracts were further purified on a glass column packed with 3 % deactivated silica gel, eluted with a dichloromethane/hexane mixture (1/1, v/v). The cleaned extracts were concentrated, solvent exchanged to methanol, and finally concentrated to 200 μL .

For water samples, the same extracts as those previously prepared for the determination of PBDEs (Wu et al. 2008) were used in this work. The extraction and cleanup procedures for water samples have been described elsewhere (Wu et al. 2008). After PBDEs and other chemical determination, the extracts were concentrated to incipient dryness under a gentle stream of nitrogen and reconstituted in 50 μL of methanol.

Fig. 1 Map of sampling sites

Internal standard (^{13}C -TBBP-A) was spiked before instrumental analysis.

Instrumental analysis

The instrumental analysis of TBBP-A was conducted according to a method previously described (Tang et al. 2015). Briefly, TBBP-A was determined using an Agilent 1200 series liquid chromatography system coupled to an Agilent 6410 triple quadrupole mass spectrometry, equipped with an electrospray interface operated in negative ionization mode (LC-ESI-MS/MS). LC separation was performed using a XDB-C₁₈ column (50 mm × 4.0 mm i.d., 1.8 μm particle size; Agilent, CA). A sample fraction volume of 3 μL was injected. The flow rate was 250 μL/min and using a gradient program consisting of two mobile phases, (A) 90 % methanol/10 % water and (B) 100 % acetonitrile. The initial mobile phase was 90 %/10 % (A)/(B) and increased to 60 %/40 % (A)/(B) over 1 min followed by an increase to 30 %/70 % (A)/(B) over 4 min, and finally followed over a 9-min period by a linear decrease to the initial condition. The target compound responses were via the multiple reaction monitoring mode with precursor ion of m/z 542.7 and product ion of m/z 79 for native TBBP-A and precursor ion of m/z 554.7 and product ion of m/z 79 for ^{13}C -labeled TBBP-A.

Quality assurance and quality control (QA/QC)

For each batch of 12 samples, a procedural blank, a spiked blank, and a pair of matrix spiked duplicate were processed. No TBBP-A was detected in the procedural blanks. The recoveries of TBBP-A spiked in the solvent and fish muscles free of TBBP-A ranged from 80 to 98 % and from 92 to 101 %, respectively. Concentrations of TBBP-A in the matrix spiking duplicate generated relative standard deviations (RSDs) less than 20 %. The reported TBBP-A concentrations were neither blank nor recovery corrected. The method detection limit (MDL), defined as a signal-to-noise ratio of 10:1, was estimated by measuring the background noise in the blank sample and peak height in the spiked sample. The MDL for TBBP-A was 10 pg/g lipid weight.

Calculations of bioaccumulation factor and trophic magnification factor

Bioaccumulation factor (BAF) was defined as the concentration ratio of the TBBP-A in biota (pg/g wet weight (ww)) to that in dissolved phase of water (pg/mL).

TL was calculated using the following relationship (Wu et al. 2009):

$$\delta^{15}\text{N} (\text{‰}) = \left[\left(R_{\text{sample}}/R_{\text{standard}} \right) - 1 \right] \times 1000 \quad (1)$$

$$\text{TL}_{\text{consumer}} = \left[\left(\delta^{15}\text{N}_{\text{consumer}} - \delta^{15}\text{N}_{\text{primary consumer}} \right) \right] / 3.4 + 2 \quad (2)$$

where R is the corresponding ratio of $^{15}\text{N}/^{14}\text{N}$ and 3.4 is the value of isotopic trophic enrichment factor. The R_{standard} was estimated using an ammonium sulfate standard.

Trophic magnification factor (TMF) was determined using the following equations (Wu et al. 2009):

$$\log \text{TBBP-A concentration (lipid-normalized)} = a + b \times \text{TL} \quad (3)$$

$$\text{TMF} = 10^b \quad (4)$$

Data analysis

For comparing BAF values between the prey fish species (mud carp and crucian carp) and the predator species (northern snakehead), the BAF data in certain fish species were firstly tested for normality using a Kolmogorov-Smirnov test. When the data was normally distributed, the two-group t test was used. Otherwise, the Mann-Whitney U test was used. Linear regression analyse was performed between the logarithm of TBBP-A concentrations and trophic level of the species. The criterion for significance that was used in all statistical tests was $p < 0.05$.

Results and discussion

TBBP-A levels

The mean concentrations of TBBP-A in target organisms from the e-waste recycling site are presented in Table 1. TBBP-A was detected in all the aquatic species, with the highest concentrations in the Chinese mystery snail (1970 pg/g ww) and the lowest concentrations in northern snakehead (350 pg/g ww). The mean TBBP-A concentration in mud carp was approximately five times higher than that in the reference mud carp samples (Table 1), suggesting that aquatic organisms from the e-waste site were heavily polluted by TBBP-A. Several types of e-waste including television or computer casings, electrical wires, and electrical components were recycled in the sampling site (Wu et al. 2008; Zheng et al. 2012). It was reported that the largest additive use of TBBP-A is in television/computer casings (Covaci et al. 2009). TBBP-A could escape from the e-waste and enter the environment during the crude recycling processes. Previous observations of TBBP-A concentrations in wild aquatic invertebrates are limited. The mean concentration of TBBP-A in our Chinese

mystery snail was eightfold higher than that (45 ng/g lw) reported in another type of mollusk, common whelk, collected from the North Sea (Morris et al. 2004). Also, the mean TBBP-A concentration in our prawn was nearly two orders of magnitude higher than those reported in another crustacean, mysid shrimp, from two sites in the Scheldt estuary, The Netherlands (0.8 and 0.9 ng/g lw) (Verslycke et al. 2005). For fish species, the current mud carp (730 pg/g ww) and northern snakehead (350 pg/g ww) showed similar concentrations with those detected in the two species (840 and 380 pg/g ww for mud carp and northern snakehead, respectively) sampled from the same e-waste recycling site as the present study (Tang et al. 2015). TBBP-A concentrations in our mud carp were also comparable to those detected in the species (mean value of 35.2 ng/g lw) from a highly industrialized area in South China (He et al. 2013). However, TBBP-A concentrations in the current fish were lower than those reported in four fish species from Lake Chaohu, China (28.5–39.4 ng/g lw) which is near TBBP-A manufacturing plants (Yang et al. 2012) and in fish from three locations (mean values of 1290, 980, and 790 pg/g ww) downstream from chemical factories or sewage treatment plants in Czech (Hloušková et al. 2013). In contrast, the TBBP-A concentrations in our fish were much higher than those detected in the edible parts of four fish species (mean values of 0.01, 0.03, 0.04, and 0.11 ng/g lw) from Japan (Ashizuka et al. 2008), the muscles of four fish species (mean value of 0.3 ng/g lw) from North Sea estuaries, The Netherlands (Morris et al. 2004), the muscles of several fish species (<0.29–1.7 ng/g lw) from English lakes (Harrad et al. 2009), and the liver of four fish species (0.3–15.4 ng/g lw) from the River Po, Italy (Luigi et al. 2015). Conclusively, TBBP-A levels detected in organisms from e-waste sites are at the higher end of the reported concentration ranges, suggesting that e-waste sites are one of the “worst-case scenarios” of TBBP-A. The elevated TBBP-A concentrations in the aquatic species from the e-waste site is of concern, because TBBP-A has been demonstrated to be toxic to aquatic organisms (Kuiper et al. 2007; Hu et al. 2015). An understanding of TBBP-A influences on these species, especially the two aquatic invertebrates, deserves further study.

Bioaccumulation potential and species-specific accumulation

To assess the bioaccumulation potential and species-specific accumulation of TBBP-A in the aquatic species, we detected TBBP-A concentrations in the dissolved phase of water (Table 1) and estimated field-determined BAFs (Fig. 2). The BAF values of 5000 and 2000 are two common regulatory thresholds for potentially bioaccumulative and highly bioaccumulative, respectively (UNEP 2001). In the present study, the mean BAF value in the Chinese mystery snail was greater than 5000, suggesting that TBBP-A is highly

Table 1 TBBP-A concentrations (mean ± SE) in dissolved phase of water (pg/L) and aquatic species (pg/g wet weight) from an e-waste recycling site and in mud carp from a reference site

	N	lipid (%)	Trophic level ^a	TBBP-A concentration
Water	6 (3) ^b	/	/	380 ± 144
The Chinese mystery snail	43 (3) ^b	0.61 ± 0.10	2.00 ± 0.04	1970 ± 560
Prawn	7 (3)	2.41 ± 0.42	2.92 ± 0.21	1570 ± 400
Mud carp	12 (8)	2.91 ± 0.35	3.07 ± 0.20	730 ± 220
Crucian carp	18 (7)	3.60 ± 0.56	2.90 ± 0.41	590 ± 230
Northern snakehead	6	1.52 ± 0.28	4.28 ± 0.17	350 ± 190
Reference mud carp	5	3.21 ± 0.25	/	145 ± 115

^aData from Wu et al. (2009)

^bNumber of individual samples collected. Figures in brackets indicate number of pooled samples when individuals were pooled

bioaccumulative in this species. A relatively high BAF value (4120) was also estimated in the prawn, which meant that TBBP-A is potentially bioaccumulative in this species. In contrast, the three fish species showed BAFs less than 2000, indicating its low bioaccumulation potential in the current fish.

The BAFs varied largely among the species, suggesting species-specific bioaccumulation of TBBP-A. Typically, BAFs were higher in the two invertebrates, i.e., the Chinese mystery snail and the prawn, relative to fish (Fig. 2). The difference in BAFs between the invertebrates and the fish is due, at least in part, to the species-specific feeding habits and the life habits and metabolic capacity for TBBP-A. The Chinese mystery snail and the prawn are detritivores, feeding on organic detritus or decomposed organic matter in the water environment (Kipp et al. 2013; Liu et al. 2005). TBBP-A is a hydrophobic organic chemical (HOC), with an octanol/water

partition coefficient (K_{OW}) of $10^{5.9}$ (European Chemicals Bureau 2006). Typically, increasing concentrations of HOCs in decomposed organic matter render the detritivores prone to higher exposure to these chemicals (deBruyn and Gobas 2004). Additionally, TBBP-A are expected to sorb onto sediment particles due to its relatively high K_{OW} . For this reason, the concentrations of TBBP-A might be higher in benthos, like snail and prawn, resulting in higher BAFs in these organisms. On the other hand, the less biotransformation and/or elimination capacities for TBBP-A in invertebrates compared to fish likely contributed to the greater BAFs in the Chinese mystery snail and the prawn. TBBP-A has been found to be biotransformed into several metabolites in fish liver subcellular fractions (Shen et al. 2012). Our previous study also suggested that the two invertebrate species investigated in the present study appeared to have lower metabolic capacities for other halogenated compounds such as PBDEs, compared to the fish species (Wu et al. 2008). For the two invertebrates, BAFs were higher in the Chinese mystery snail (mean = 5190) than in the prawn (mean = 4120). The Chinese mystery snail is a benthic dwelling organism and uses a pulmonary respiration, only remains near the water surface when filling the lung (Kipp et al. 2013). In contrast, the prawn is an epibenthic crustacean and is fully submerged throughout the exposure, using gill respiration (Liu et al. 2005). It is suggested that respiration strategy and locomotion mode may largely influence the bioaccumulation of contaminants in aquatic species (Borgå et al. 2004; Rubach et al. 2011), although there is no such data on TBBP-A. Comparing BAFs between the prey fish species and the predator fish, mud carp (two group *t* test, $p=0.003$) and crucian carp (two group *t* test, $p=0.001$) showed significantly higher BAFs than northern snakehead. The result is inconsistent with that observed for other brominated flame retardants such as PBDEs which showed higher BAFs in predator fish than prey fish (Wu et al. 2008). The lower BAFs estimated in northern snakehead is possibly due to the greater metabolism capacity and/or the less assimilation efficiency for TBBP-A in this species compared to the other

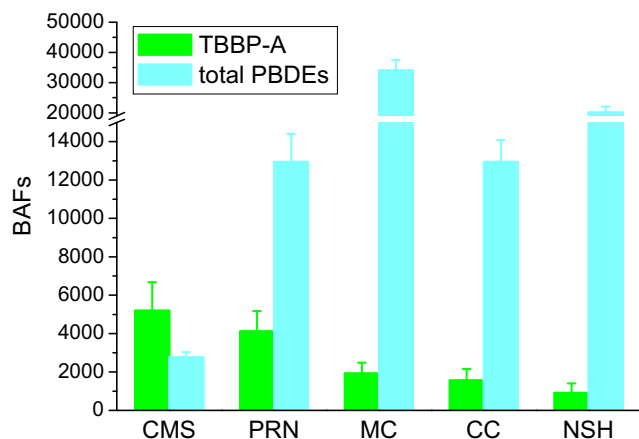


Fig. 2 Field-determined bioaccumulation factors (BAFs) of TBBP-A in the aquatic species from an e-waste recycling site in South China, with comparative data for total PBDEs (the sum concentrations of BDEs 28, 47, 99, 100, 138, 153, 154, and 209). BAF data for PBDEs is from Wu et al. (2008). Error bars represent ±1SE. CMS the Chinese mystery snail (*Cipangopaludina chinensis*), PRN the prawn (*Macrobrachium nipponens*), MC mud carp (*Cirrhinus molitorella*), CC crucian carp (*Carassius auratus*), NSH northern snakehead (*Ophicephalus argus*)

two fish species. A recent study also reported higher TBBP-A concentrations in mud carp than northern snakehead collected from an e-waste recycling site in South China (Tang et al. 2015).

The BAFs for Σ PBDEs (sum concentrations of BDEs 28, 47, 99, 100, 138, 153, 154, and 209) have been previously estimated in the same sample set as the present study (Wu et al. 2008). In the Chinese mystery snail, the mean BAF for TBBP-A is twofold greater than that for Σ PBDEs (Fig. 2), suggesting higher bioaccumulation potential for TBBP-A compared to Σ PBDEs in this species. However, the BAFs for TBBP-A in the other species investigated were 3–22 times lower than those estimated for Σ PBDEs (Fig. 2). We hypothesize that this is because the greater metabolism or elimination capacities for TBBP-A than PBDEs in these species.

Reports on BAFs for TBBP-A are limited. Our BAFs estimated in the fish species were comparable to those (400–39800) calculated in three fish species from a river in a highly industrialized area in South China (He et al. 2013), but were much higher than that (0.62) calculated in fish from English lakes (Harrad et al. 2009), and those (0.53–0.84) in healthy scallops from the Yellow Sea, China (Hu et al. 2015). The species-specific accumulation of TBBP-A and the site-specific environmental TBBP-A concentrations among these studies may account for the observed differences in BAF for TBBP-A. It should be noted that the estimated BAFs for TBBP-A were generally higher in the e-waste recycling sites and the industrialized areas, compared to other sampling sites. Such exposure scenarios challenge the risk assessment of TBBP-A conducted by the European Union (European Chemicals Bureau 2008). The high bioaccumulation potentials for TBBP-A in the aquatic invertebrates from the worst-case scenarios is of concern, because TBBP-A has been demonstrated to be toxic to these animals (Sverdrup et al. 2006; Hu et al. 2015). An advanced understanding of mechanisms responsible for aquatic invertebrates bioaccumulation of TBBP-A is needed.

Trophic transfer along the aquatic food web

We have previously determined the TL and constituted the food web structure of the current aquatic species (Table 1), using stable nitrogen isotope composition ($\delta^{15}\text{N}$) in these species (Wu et al. 2009). To further assess the trophic transfer of TBBP-A in the current food web, we investigated the relationship between the lipid-normalized concentrations of TBBP-A and the TLs of the species, and calculated TMF of TBBP-A in the food web. As shown in Fig. 3, TBBP-A concentrations were negatively correlated to the TLs of the species ($r = -0.82$), although this correlation was not statistically significant ($p = 0.09$). The result suggests that TBBP-A experienced

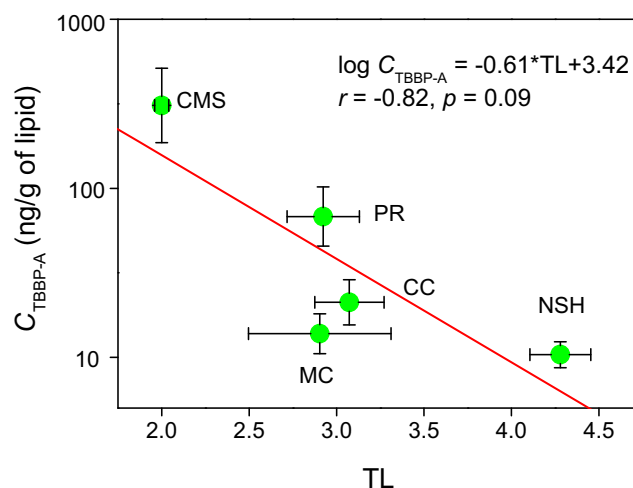


Fig. 3 Relationship between TBBP-A concentrations and trophic level (TL) of the aquatic species from an e-waste recycling site in South China. Error bars represent $\pm 1\text{SE}$. CMS the Chinese mystery snail (*Cipangopaludina chinensis*), PRN the prawn (*Macrobrachium nipponense*) MC mud carp (*Cirrhinus molitorella*), CC crucian carp (*Carassius auratus*) NSH northern snakehead (*Ophicephalus argus*)

trophic dilution in the current food web. The estimated TMF was 0.25, meaning that TBBP-A concentrations decreased approximately 4-fold in average after a trophic level transfer. Trophic dilution of some other non-PBDE brominated flame retardants have been previously reported in the same food web as the current study (Wu et al. 2010). However, food-web magnification of some PBDE congeners (TMFs = 1.35–4.47) has been observed in the same food web (Wu et al. 2009).

When trophic dilution is observed for hydrophobic chemicals, one possible reason could be substantial biotransformation rates and/or poor assimilation efficiencies in species at higher TLs (Wan et al. 2010). Biotransformation of TBBP-A in fish species have been reported previously (Shen et al. 2012), although it is occurred in different species from our fish. Besides biotransformation and assimilation efficiencies, the relative importance of exposure routes other than dietary uptake, e.g., respiratory uptake, in organisms at lower TLs could also result in trophic dilution of a chemical (Borgå et al. 2004, 2012). When assessing the TMF for a chemical, it is presumed that the major exposure route for a species is through its diet. However, for aquatic species that take up chemicals through other exposure pathways such as respiratory surfaces can highly likely affect the determination of TMF in an aquatic food web (Borgå et al. 2004, 2012). Although there is, to our knowledge, no such a study demonstrating the relative fractions of respiratory uptake of TBBP-A in aquatic species, it is likely occurring because of the relatively high water solubility of TBBP-A (63 $\mu\text{g/l}$) (European Chemicals Bureau 2008). If respiratory exchange (inhalation) does occur in aquatic organisms, it may be more pronounced for lower TL species with high surface area to body ratios (Borgå et al.

2004, 2012), resulting in a TMF less than 1 (trophic dilution in the food web).

Conclusions

In conclusion, TBBPA was detected in several wild aquatic species collected from an e-waste recycling site in South China, at higher concentrations compared with most of what has been reported for wild aquatic species. Although TBBP-A experienced trophic dilution in the present food web, field determined BAFs suggested that TBBP-A is bioaccumulative in the low trophic level organisms. Since the toxicity of TBBPA and the highly bioaccumulative characteristic in low trophic level species, more monitoring to examine the levels of TBBP-A in the environment of e-waste recycling sites and its effects on low trophic level wildlife would assist at better understanding the fate and the effects of this chemical.

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