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Occurrence and trophic magnification profile of triphenyltin compounds in marine mammals and their corresponding food webs

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ABSTRACT

The occurrence of triphenyltin (TPT) compounds, a highly toxic antifouling biocide, has been documented in marine environments and organisms all over the world. While some studies showed that marine mammals can be used as sentinel organisms to evaluate the pollution status of emerging contaminants in the environment because of their long lifespans and high trophic levels, information regarding the contamination status of TPT in marine mammal species has been limited over the past decade. More importantly, the primary bioaccumulation pathway of TPT in these long-lived apex predators and the corresponding marine food web is still uncertain. Therefore, this study aimed to evaluate the contamination statuses of TPT in two marine mammal species, namely the finless porpoise and the Indo-Pacific humpback dolphin, and assess the trophic magnification potential of TPT along the food webs of these two species, using stable isotope analysis, and chemical analysis with gas chromatography-mass spectrometry. The results showed that TPT is the predominant residue in majority of the analyzed individuals of two marine mammals, with concentrations ranging from 426.2 to 3476.6 ng/g wet weight in their muscle tissues. Our results also demonstrated an exponential increase in the concentration of TPT along the marine food web, indicating that trophic magnification occurs in the respective food webs of the two marine mammals. The range of trophic magnification factors of TPT in the food webs of finless porpoise and Indo-Pacific humpback dolphin was 2.51–3.47 and 2.45–3.39, respectively. These results suggest that high trophic organisms may be more vulnerable to the exposure of TPT-contaminated environments due to the high trophic magnification potential, and thus ecological risk of these compounds ought to be assessed with the consideration of their bioaccumulation potentials in these marine mammals.

1. Introduction

Emerging environmental contaminants can be readily bioaccumulated in organisms through various routes, resulting in a higher concentration in the organisms than their surrounding environment (Gobas and Morrison, 2000). Such a phenomenon is especially profound in predatory marine mammals because of their long lifespans and high trophic levels. Therefore, these organisms are widely acknowledged as sinks for emerging contaminants to assess the status of environmental pollution in the marine environment (Zhu et al., 2014), and sentinel organisms to evaluate the ecosystem's health (Aguirre and Tabor, 2004).

The widespread of organotin compounds, especially triphenyltin (TPT) compounds, in coastal areas around the world has recently raised many concerns because of their high toxicities to a wide range of marine organisms (Yi et al., 2012). TPT compounds are renowned endocrine disruptors, which can lead to the development of male sexual characteristics in female gastropods when exposed to TPT at environmentally realistic concentrations (Horiguchi et al., 1997; Yi et al., 2012; Ho et al., 2016). These compounds can also cause immunomodulatory effects to marine mammals, which would consequently increase their susceptibility to virus-associated infectious diseases (Nakayama et al., 2009), and eventually lead to virus-associated mass mortality events (van Loveren et al., 2000).

The contamination status of TPT in environmental compartments such as seawater and marine sediment have been comprehensively investigated in different parts of the world (e.g. Yi et al., 2012). Yet, the

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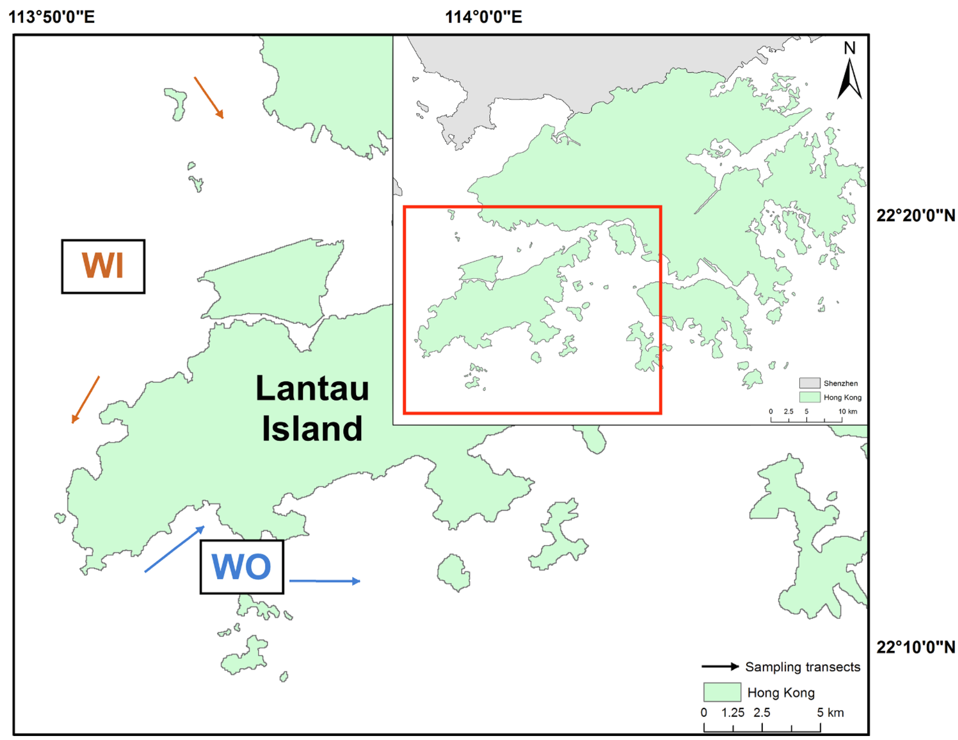

Fig. 1. Map showing the sampling locations: the insert is a map of Hong Kong and the enlarged figure shows the sampling transects in Inner Estuary (west Lantau, WI) and Outer Estuary (south Lantau, WO) of the western waters of Hong Kong.

data on marine mammals are relatively lacking. For example, the latest figure on TPT levels in the finless porpoises *Neophocaena phocaenoides* inhabiting Hong Kong and its adjacent waters can be dated back to 2003 (Nakayama et al., 2009), whereas no record of TPT contamination can be found in the local population of Indo-Pacific humpback dolphins *Sousa chinensis* (Sanganyado et al., 2018). These resident marine mammal species are of high conservation and protection concerns because they are listed in Appendix A of the Convention of International Trade in Endangered Species of Wild Fauna and Flora (CITES) (Hung et al., 2007) and categorized as "Vulnerable" in the International Union for Conservation of Nature (IUCN) Red List (Jefferson et al., 2017; Wang and Reeves, 2017). More importantly, the local marine mammals in of Hong Kong showed higher neonatal mortality rates after exposure to immunosuppressing organic contaminants because they compromised their immune functions upon exposure (Parsons and Chan, 1998). Therefore, there is an apparent need to assess the level of contamination of TPT and its degradation compounds (mono- and di-phenyltin; MPT and DPT), collectively known as phenyltin compounds (PTs), in the marine mammals, so as to evaluate their ecological risks and carry out consequent conservation plans to protect these species, especially in highly urbanized megacities like Hong Kong which was subjected to the serious TPT contamination (Yi et al., 2012; Ho et al., 2016).

Furthermore, biomagnification of TPT has been demonstrated in the lower part of the marine food chain (i.e., planktonic organisms, invertebrates, and fishes) (Hu et al., 2006b; Kono et al., 2008; Murai et al., 2008). However, the potential of this compound to biomagnify in the organisms at higher trophic levels, especially in air-breathing organisms such as marine mammals and seabirds, remains debatable. To date, only two documented studies included marine mammals or seabirds as the apex predators in the investigation of biomagnification of TPT. Strand and Jacobsen (2005) reported that marine mammals in Danish coastal waters generally had lower TPT concentrations than the analyzed invertebrate and fish species; while He et al. (2018) demonstrated that seals and seabirds had the highest concentrations of TPT in comparison to the other organisms in the same food web. The lack of studies and the discrepancy of results urge additional studies to assess the biomagnification potential of TPT compounds in marine food webs with mammals as apex predators so as to provide sufficient clues for further evaluation of environmental risks associated with TPT compounds. Therefore, the current study was specially designed to reveal the biomagnification potential of TPT in marine organisms, especially covering marine mammals which occupy high trophic levels of the marine food web. In particular, the trophic magnification factor (TMF), which is the average measurement of biomagnification throughout a food web (Borgå et al., 2012), was adopted for the present investigation. Such an approach has been widely recognized as one of the most holistic measures in characterizing bioaccumulation because it accounts for the trophic interaction and transfer of contaminants within a food web (Borgå et al., 2004).

Based on the existing environmental concentrations of TPT recorded in the western waters of Hong Kong (Ho et al., 2016; Sham, 2019), it is hypothesized that the marine mammals residing this area are highly contaminated with TPT, with dietary transfer being one of the most significant pathways of the uptake of these contaminants. On the whole, this study aimed to (1) evaluate the contamination status of PTs in marine mammals inhabiting Hong Kong, i.e., the Indo-Pacific humpback dolphin (*S. chinensis*) and the finless porpoise (*N. phocaenoides*), and (2) characterize the trophic magnification profile of the three PT residues in the coastal marine food web of Hong Kong, in particular in organisms occupying high trophic positions. Results obtained from this study will be essential for the assessment and management of ecological risks of these widespread contaminants in the marine environment of Hong Kong and the nearby waters, with the consideration of the contamination status of TPT in marine mammals and along their food webs.

2. Materials and methods

2.1. Sampling sites

Sampling of organisms was conducted at two sites of the western waters of Hong Kong, namely the Inner Estuary (WI) and Outer Estuary (WO) (Fig. 1), which are the main habitats of the two resident marine mammal species in Hong Kong (Hung, 2018). The Indo-Pacific humpback dolphin and the finless porpoise show segregation in habitat use, of which the dolphins are generally found in the more estuarine habitat in WI, while finless porpoises are found to inhabit the more oceanic, saline water in WO and southern waters of Hong Kong (Hung, 2018). Both of these species are known to be the apex predator of their respective food webs (Hung et al., 2006b; Gui et al., 2014).

2.2. Sample collection and preparations

Five individuals of Indo-Pacific humpback dolphins (*S. chinensis*) and eight individuals of finless porpoises (*N. phocaenoides*) were used as target organisms in this study. Muscle samples of all marine mammal individuals were obtained during 2016–2017 in collaboration with the Ocean Park Conservation Foundation Hong Kong (OPCFHK), which collected stranded individuals and recorded their biological information (e.g. total length, sex, and maturity stage; see Table 1) following the description in the OPCF standard operation protocol (OPCF, 2016) developed based on Jefferson et al. (2012).

Furthermore, five species of mollusks, three crustaceans, and twelve fishes (Table 2) were collected during the dry season (October–December) of 2015 by a shrimp trawler with a scientific permit from the Agriculture Fisheries and Conservation Department of the Government of the Hong Kong Special Administrative Region (permit no. R1710007). These species were selected based on their relative abundance in the western waters of Hong Kong, as well as their relative trophic positions in the selected food webs. More importantly, multiple selected fish species, such as *Johnius* spp. and *Collichthys lucidus*, were reported to be the major preys of the Indo-Pacific humpback dolphins (Barros et al., 2004).

Muscle tissues were used for the both quantification of PT compounds and determination of trophic level. All dissected muscle samples were freeze-dried (VirTis #6KBTES-55 freeze dryer, Gardiner, NY, U.S.A.) for at least 48 h to attain a constant dry weight, and homogenized with a blender (Philips HR2860, Holland) for subsequent analyses.

2.3. PT analyses

PT compounds were extracted following the protocols from Xie et al. (2010). In brief, the target compounds were extracted from the dried samples using toluene and glacial acetic acid mixture (toluene:AcOH = 10:4; Tedia Company, OH, USA) and 0.5% ammonium pyrrolidinedithiocarbamate solution (J&K Scientific, Beijing, China), followed by derivatization with Grignard reagents (2 M pentylmagnesium bromide in diethyl ether; J&K Scientific, Beijing, China) and clean-up with a column packed with Florisil (Acros Organics, Thermo Fisher Scientific, MA, USA) and anhydrous sodium sulfate (Sinopharm Chemical Reagent, Shanghai, China). Elutes from Florisil clean-up were then concentrated with a stream of nitrogen until a final volume of 0.1 mL was obtained. The resulting solutions were transferred to amber vials for quantification using gas-chromatography (GC; Agilent 6890) equipped with a mass-selective detector (MS; Agilent 5973). The average recovery of the quantification method was 90.3%. All concentrations were converted to ng/g wet weight (w.w.) using conversion factors obtained through previous experiments with dissected samples or from literatures (Supplementary Table S1).

2.4. Food web characterization and trophic level determination

Only dolphins that reached juvenile stage or above were used in the analyses of food web and trophic level. New-born dolphins, i.e., the "neonate/calf" group, were excluded because these individuals were known to feed on milk from their mothers. Stable carbon (C) and nitrogen (N) isotopes of marine mammals and other organisms were measured using the Nu Perspective Isotope Ratio Mass Spectrometer (Nu Instrument Ltd., Wrexham, U.K.) coupled with an Elemental Analyzer (Eurovector EA 3028, Isomass Scientific Inc., Alberta, Canada) at the Stable Isotope Laboratory of the University of Hong Kong (Central Facility of Science Faculty). To eliminate the variation in $\delta^{13}C$ caused by excessive lipids, normalization was performed if the C:N of samples was greater than 3 using the equation below (Mak, 2017; Perkins et al., 2018; Post et al., 2007):

$$\delta^{13}C_{normalized} = C: N \times 1.1124 + \delta^{13}C_{original} - 2.9802 (R2 = 0.899)$$

Trophic levels (TL) of organisms were calculated using stable nitrogen isotope ($\delta^{15}N$) as a proxy. Since the isotopic fractionations in the selected food webs were not well-established, we adopted two widely-applied trophic enrichment factors (TEF) for $\delta^{15}N$, 2.5‰ and 3.4‰, to estimate the trophic position of the sampled organisms in order to provide a conservative range of $\delta^{15}N$, as well as TL for the calculations of TMF (Wai et al., 2011; Borgå et al., 2012). The former value (2.5‰) was derived from the coastal marine ecosystem of Hong Kong in a previous study (Wai et al., 2011), while the latter value (3.4‰) is a

Table 1

Results of stable isotope analysis and quantification of each phenyltin (PT) compound (i.e., mono-, di-, and tri-phenyltin; MPT, DPT, and TPT) and total PT in tissues of the marine mammals. Biological measurements (i.e., sex, length and maturity stage), stable isotope data ($\delta^{13}C$ and $\delta^{15}N$), estimated trophic levels (TL), and concentrations of various PT compounds in each individual marine mammal are shown. ND means that the data were not determined. The range of TL values were calculated using two different TEFs as described in the Materials and Methods.

Individual ID	Sex	Length (cm)	Maturity stage	$\delta^{13}C$ (‰)	$\delta^{15}N$ (‰)	TL	MPT Conc. (ng/g w.w.)	%	DPT Conc. (ng/g w.w.)	%	TPT Conc. (ng/g w.w.)	%	Total PT Conc. (ng/g w.w.)
Finless porpoise (*Neophocaena phocaenoides*)													
NP17–0705	F	73	Juvenile	−14.92	14.61	3.80–4.45	688.0	37.0	339.6	18.3	830.6	44.7	1858.2
NP17–3101	M	127	Juvenile	−13.84	13.89	3.59–4.17	2636.9	42.0	187.9	3.0	3455.6	55.0	6280.4
NP17–1503	M	138	Juvenile	−13.47	15.64	4.11–4.86	167.3	12.3	244.1	18.0	945.4	69.7	1356.7
NP16–2209	M	150	Sub–adult	−14.60	16.83	4.46–5.34	10.1	0.8	65.5	5.1	1218.1	94.2	1293.7
NP16–2709	M	152	Sub–adult	−15.54	15.53	4.07–4.82	12.7	0.9	110.8	8.2	1233.6	90.9	1357.1
NP17–1012	F	141	Adult	−14.19	14.31	3.72–4.33	125.5	13.9	201.9	22.4	573.2	63.7	900.5
NP17–1403	M	160	Adult	−14.56	14.74	3.84–4.05	990.9	32.3	515.3	16.8	1559.3	50.9	3065.4
NP17–2203a	M	170	Adult	−14.25	15.42	4.04–4.78	146.8	6.5	107.0	4.7	2004.9	88.8	2258.7
Indo–Pacific humpback dolphins (*Sousa chinensis*)													
SC17–1408	F	107	Neonate/calf	ND	ND	ND	475.5	35.0	152.0	11.2	732.3	53.9	1359.8
SC17–0506	U	110	Neonate/calf	ND	ND	ND	1978.2	70.8	391.1	14.0	426.2	15.3	2795.5
SC16–2306a	M	115	Neonate/calf	ND	ND	ND	72.9	4.3	126.1	7.4	1503.4	88.3	1702.3
SC17–0601	U	269	Adult	−16.49	18.34	4.79–5.80	< 0.1	< 0.1	28289.0	89.5	3330.2	10.5	31619.2
SC16–2306b	M	272.5	Adult	−18.05	21.34	5.68–7.00	689.9	15.8	192.0	4.4	3476.6	79.8	4358.4

* Sex: M = Male; F = Female; U = Unknown.

Table 2
Trophic information of each species analyzed in this study from respective sampling sites (i.e., Inner Estuary (WI) and Outer Estuary (WO) in the western waters of Hong Kong), including stable isotope signatures in their tissues ($\delta^{13}C$ and $\delta^{15}N$) and estimated trophic levels (TL). *n* represents number of replicates used for stable isotope analysis, while N/A means that samples were not available. The average values of $\delta^{13}C$ and $\delta^{15}N$ are shown while numbers in brackets represent standard deviations. The range of TL values were calculated using two different TEFs.

Taxon	Species name	WI				WO			
		n	δ13C (‰)	δ15N (‰)	TL	*n*	δ13C (‰)	δ15N (‰)	TL
Mollusks	*Amphioctopus fangsiao*	3	−16.09 (0.32)	12.45 (0.25)	3.06–3.44	N/A	N/A	N/A	N/A
	Fulvia australis	N/A	N/A	N/A	N/A	3	−16.80 (0.10)	7.67 (0.17)	1.67–1.76
	Nassarius siquijorensis	3	−18.32 (0.17)	12.94 (0.73)	3.21–3.64	3	−15.75 (0.26)	11.65 (1.10)	2.93–3.27
	Pinctada imbricata fucata	3	−19.28 (0.22)	9.09 (0.40)	2.07–2.10	3	−18.27 (0.75)	8.65 (0.25)	2.05–2.07
	Turritella bacillum	3	−18.36 (0.23)	9.13 (0.07)	2.09–2.12	3	−16.16 (0.27)	8.36 (0.08)	1.95–1.96
Crustaceans	*Charybdis feriata*	3	−17.23 (0.35)	12.27 (0.81)	3.01–3.37	3	−15.92 (0.19)	11.53 (0.24)	2.90–3.22
	Harpiosquilla harpax	3	−18.09 (0.07)	14.24 (0.13)	3.59–4.16	3	−16.51 (0.21)	12.58 (0.14)	3.21–3.64
	Oratosquilla oratoria	3	−18.62 (0.35)	13.10 (0.74)	3.25–3.70	3	−15.74 (0.05)	10.99 (0.49)	2.74–3.00
Fishes	*Chrysochir aureus*	3	−17.85 (0.00)	14.89 (0.00)	3.78–4.42	N/A	N/A	N/A	N/A
	Collichthys lucidus	3	−18.42 (0.73)	12.96 (0.39)	3.21–3.65	N/A	N/A	N/A	N/A
	Cynoglossus bilineatus	3	−18.77 (3.99)	14.03 (0.38)	3.53–4.07	N/A	N/A	N/A	N/A
	Dasyatis zugei	3	−18.63 (0.54)	14.46 (0.62)	3.65–4.25	N/A	N/A	N/A	N/A
	Johnius belangerii	3	−17.90 (2.25)	14.50 (1.28)	3.66–4.26	N/A	N/A	N/A	N/A
	Johnius heterolepis	3	−16.42 (1.12)	13.60 (0.66)	3.40–3.90	N/A	N/A	N/A	N/A
	Pisodonophis cancrivorus	N/A	N/A	N/A	N/A	3	−15.42 (0.12)	13.92 (0.56)	3.60–4.17
	Platycephalus indicus	3	−19.67 (1.55)	15.55 (1.52)	3.97–4.69	3	−17.27 (0.93)	13.64 (0.67)	3.52–4.07
	Scoliodon laticaudus	3	−19.00 (0.84)	16.02 (0.35)	4.11–4.87	N/A	N/A	N/A	N/A
	Siganus canaliculatus	N/A	N/A	N/A	N/A	3	−15.61 (0.68)	12.08 (0.74)	3.06–3.44
	Takifugu ocellatus	N/A	N/A	N/A	N/A	3	−18.80 (1.89)	14.77 (1.34)	3.85–4.51
	Trypauchen vagina	N/A	N/A	N/A	N/A	3	−16.93 (0.15)	12.99 (0.23)	3.33–3.80

universal value commonly adopted in marine trophic studies worldwide (Borgå et al., 2012). The equation for calculating TL is shown as follows:

$$TL_{\text{consumer}} = 2 + \frac{(\delta^{15}N_{consumer} - \delta^{15}N_{baseline})}{\text{TEF}}$$

The $\delta^{15}N$ baseline values used for TL calculation were 8.84 in WI and 8.48 in WO (Tao, 2018). These values were obtained by averaging all known filter-feeders collected in 2015 from the respective sites (Post, 2002).

2.5. TMF calculation

To assess whether organisms can bioaccumulate PT and each corresponding residue (i.e., MPT, DPT, and TPT) from their diets, TMF was adopted to evaluate the biomagnification potential of these compounds. Regression analyses were performed with TL as independent variables, and log-transformed concentration of each PT residue as dependent variables. The TMF was further calculated using the slope of the equation if the regression was found to be significant ($p < 0.05$). Detailed equations used are shown below (Borgå et al., 2012):

$$\log_{10}[TPT] = a + b \times TL$$

$$\text{TMF} = 10^b$$

Concentrations of PTs were not undergone normalization to lipid contents since PT compounds are found to have higher affinity to proteins than to lipids (Rose and Aldridge, 1968; Strand and Jacobsen, 2005; Sham, 2019).

2.6. Statistical analyses

The differences in TPT concentrations between the two marine mammal species were tested using Student's independent sample *t* test if data were normally distributed. The same statistical test was applied to compare the differences between the average $\delta^{13}C$ and $\delta^{15}N$ after correcting to the respective baseline values of the two analyzed sites ($\alpha = 0.05$). Linear regression analysis was employed to evaluate the relationship between the tropical level and the tissue concentration of each target chemical contaminant.

3. Results

3.1. PT contamination in marine mammals

All three PTs were detected in all marine mammal samples, with greatly different composition profiles of PTs across individuals (Table 1). TPT contributed 10.5–94.2% to total PTs, whereas DPT and MPT contributed 3.0–89.5% and < 0.1–70.8%, respectively. TPT was always the predominant compound in the eight finless porpoises (*N. phocaenoides*), while TPT was dominant in only three out of five individuals of Indo-Pacific humpback dolphins (*S. chinensis*).

Nonetheless, the contamination status of TPT in marine mammals is of high concern in this study. Tissue concentrations of TPT in the analyzed marine mammal individuals varied from 426.2 to 3476.6 ng/g wet weight (w.w.) (Table 1). The mean TPT concentration in Indo-Pacific humpback dolphins was 1893.8 ng/g w.w., while the average concentration of TPT in finless porpoises was 1477.6 ng/g w.w. No statistical difference of the mean TPT concentrations was identified between the two species ($t_{0.05(2),\ 6} = -0.580$, $p = 0.583$).

No statistical analyses were carried out to identify differences in TPT contamination status between sex, and among maturity stages due to the insufficient sample size. In general, the concentration of TPT increased as dolphins and porpoises matured, with an exception found in one juvenile finless porpoise individual, which contained the highest TPT concentration among all individuals of the finless porpoise samples (3455.6 ng/g w.w.) (Table 1). Males, in general, showed a higher concentration of TPT than females in both species (Table 1).

3.2. Food web structure of selected sites

A clear separation in community isotopic niche between WI and WO was shown in the biplot of $\delta^{13}C$ against $\delta^{15}N$ (Fig. 2). The $\delta^{13}C$ in WI and WO differed significantly ($t_{0.05(2),\ 34} = -5.846$, $p < 0.001$), which more depleted $\delta^{13}C$ values were found in WI (range: −19.67 to −16.09‰), whilst more enriched $\delta^{13}C$ values were found in WO (range: −18.80 to −13.47‰) (Tables 1 & 2). However, no significant difference was detected in $\delta^{15}N$ between the two sites ($t_{0.05(2),32} = 1.667$, $p = 0.105$). The range of $\delta^{15}N$ of the analyzed biota was 9.09–21.34‰ in WI and 7.67–16.83‰ in WO, respectively (Tables 1 & 2). A range of

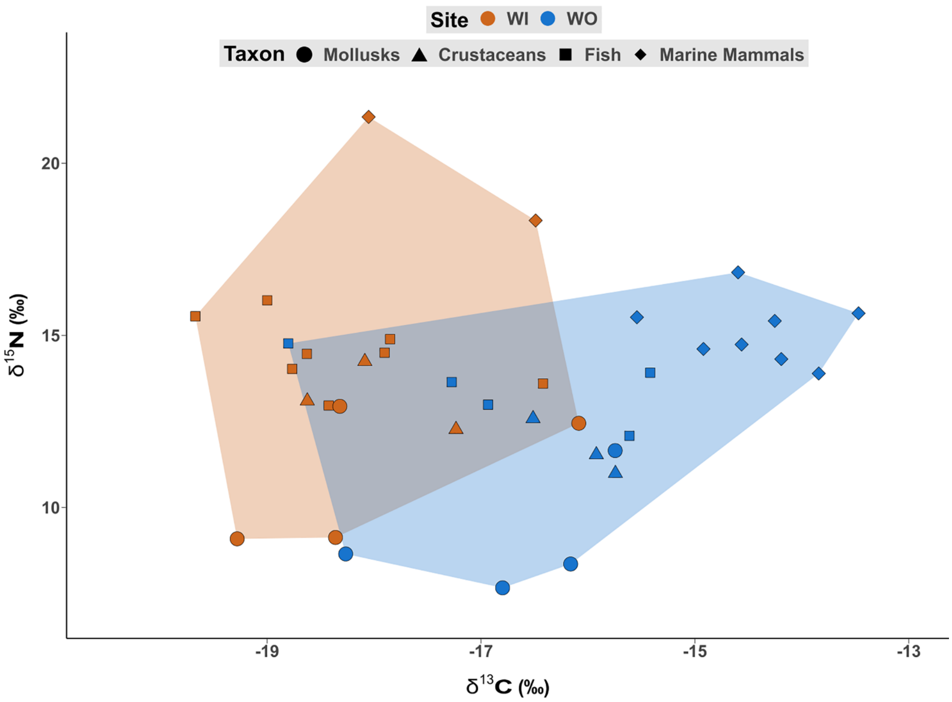

Fig. 2. Bi-plot of $\delta^{13}C$ and $\delta^{15}N$ signatures in muscle tissues of various marine organisms (mollusks, crustaceans, fishes and marine mammals) collected at Inner Estuary (WI) and Outer Estuary (WO) in the western waters of Hong Kong.

trophic level (TL) of each organism was calculated and shown in Tables 1 and 2 for organisms at low and high trophic levels, respectively. The lowest and highest TLs in WI were 2.07 and 7.00, respectively, whereas TL in WO ranged from 1.67 to 5.34.

3.3. *Trophic transfer of PT residues*

Concentrations of all PT residues in water-respiring organisms, including mollusks, crustaceans, and fishes, are presented in Table 3. In the food web of the Indo-Pacific humpback dolphins (WI), concentrations of all PT residues except MPT were found to increase exponentially with TL (Table 4; Fig. 3). TMF of each compound was calculated from the slope of each significant regression. DPT was found to have the largest TMF of 6.03–11.48 among the four analyzed groups of compounds, while TPT had a TMF of 2.45–3.39 (Table 4; Fig. 3).

Trophic magnification profiles in the food web of the finless porpoise (WO) were different from that of WI. Among the three PT residues, only concentration of TPT was found to have a significantly positive relationship with TL, with a TMF of 2.51–3.47 (Table 4; Fig. 4). Total PT concentration also significantly increased with increasing TL (p = 0.050). MPT and DPT did not increase significantly with TL (Table 4; Fig. 4).

4. Discussion

4.1. *PT contamination in marine mammals inhabiting Hong Kong waters*

The current study adopted marine mammals as sentinel organisms to assess the contamination status of PTs in Hong Kong waters. TPT was the dominant PT residue in muscle tissues of finless porpoises (*N. phocaenoides*), but only three out of five Indo-Pacific humpback dolphins (*S. chinensis*) followed this trend. Results of finless porpoises agreed well with previous studies that TPT was the dominant PT residue in marine mammal tissues (Supplementary Table S2). In contrast, the relatively large amount of degradation products (i.e., MPT and DPT) detected in Indo-Pacific humpback dolphins may be attributed to the biodegradation of TPT in their body because some marine cetaceans have been found to contain detoxification enzymes that are responsible for the degradation of TPT (Goksøyr, 1995; Ohhira et al., 2006). In addition, prey items of the studied marine mammals inhabiting the same area were also found to be highly contaminated by DPT and MPT (Sham, 2019), which could also lead to high concentrations of these compounds in the dolphins through dietary transfer (see Section 4.4).

In the present study, the highest tissue concentration of TPT was identified in an adult individual of Indo-Pacific humpback dolphins (3476.6 ng/g w.w.), which was approximately five times higher than the highest TPT concentration ever documented in marine mammals worldwide (false killer whale: 694.0 ng/g w.w.; Harino et al., 2007a; Supplementary Table S2). In contrast to the high concentrations of TPT recorded in this study (Indo Pacific humpback dolphins' range: 426.2–3476.6 ng/g w.w.; finless porpoises' range: 573.2–3455.6 ng/g w.w.), the populations of the same mammal species in Thailand were reported to contain no more than 1 ng/g w.w. of TPT in their tissues (Harino et al., 2007b). Nakayama et al. (2009) also distinguished Hong Kong as the contamination "hotspot" of TPT because concentrations of TPT in the finless porpoise population collected from Hong Kong waters in 2003 were found to be 10 times higher than that in the populations in Japan, ranging from 200 to 400 ng/g w.w. in their livers. These observations suggest that marine mammals inhabiting Hong Kong waters are severely contaminated by TPT in comparison to other marine habitats around the world.

Conspicuously, the current study recorded a drastic increase in TPT contamination in marine mammals in Hong Kong over the past decade in comparison to the values documented in previous studies. The highest concentration of 3455.6 ng/g w.w. in the muscles of the finless porpoises detected in this study was approximately 10 times higher than the highest value recorded in 2003 by Nakayama et al. (2009). A similar increasing trend of PT contamination was also revealed by a long-term (1990–2015) biomonitoring of PT contamination in the marine gastropod *Reishia clavigera* in the marine environment of Hong Kong (Ho et al., 2016). These results jointly indicated that marine organisms inhabiting Hong Kong waters have become more severely contaminated with PTs over the past 10 years, which could be ascribed to the delayed adoption of OT-based antifouling paint restrictions in

Table 3
Average concentrations (standard deviations in brackets) of each phenyltin (PT) compound (i.e., mono-, di-, and tri-phenyltin; MPT, DPT, and TPT) and total PT in muscle tissues of marine organisms. N/A means sample not available.

Taxon	Species name	WI				WO			
		MPT (ng/g w.w.)	DPT (ng/g w.w.)	TPT (ng/g w.w.)	Total PT (ng/g w.w.)	MPT (ng/g w.w.)	DPT (ng/g w.w.)	TPT (ng/g w.w.)	Total PT (ng/g w.w.)
Mollusks	*Amphioctopus fangsiao*	0.0 (0.0)	39.5 (36.0)	185.4 (69.2)	224.9 (105.1)	N/A	N/A	N/A	N/A
	Fulvia australis	N/A	N/A	N/A	N/A	461.4 (458.9)	281.3 (306.3)	230.2 (159.3)	972.9 (920.1)
	Nassarius siquijorensis	0.0 (0.0)	0.0 (0.0)	142.7 (19.0)	142.7 (19.0)	1597.3 (1108.5)	530.5 (551.6)	88.8 (9.8)	2216.7 (1460.4)
	Pinctada imbricata fucata	21.1 (36.6)	23.8 (41.1)	81.0 (28.2)	125.9 (31.2)	0.0 (0.0)	0.0 (0.0)	93.5 (10.1)	93.5 (10.1)
	Turritella bacillum	0.0 (0.0)	0.0 (0.0)	104.5 (27.7)	104.5 (27.7)	554.3 (368.0)	153.7 (95.1)	54.7 (8.9)	762.6 (471.0)
Crustaceans	*Charybdis feriata*	0.0 (0.0)	19.9 (17.2)	166.7 (45.2)	351.8 (140.3)	0.7 (1.3)	13.5 (11.3)	61.3 (37.1)	75.5 (49.6)
	Harpiosquilla harpax	123.0 (176.7)	62.1 (8.8)	166.7 (45.2)	351.8 (140.3)	8.7 (5.8)	45.7 (4.4)	205.7 (21.6)	260.1 (31.7)
	Oratosquilla oratoria	17.6 (1.4)	47.9 (9.6)	169.3 (50.3)	17.6 (59.7)	6.4 (5.7)	32.5 (3.5)	173.5 (9.2)	212.5 (17.4)
Fishes	*Chrysochir aureus*	101.3 (101.4)	128.8 (75.7)	684.5 (362.6)	914.6 (426.8)	N/A	N/A	N/A	N/A
	Collichthys lucidus	3.4 (0.3)	1.2 (0.2)	261.0 (27.0)	265.6 (27.3)	N/A	N/A	N/A	N/A
	Cynoglossus bilineatus	7.6 (3.1)	1.6 (0.3)	485.6 (97.5)	494.8 (95.0)	N/A	N/A	N/A	N/A
	Dasyatis zugei	18.0 (31.2)	6.0 (3.0)	210.2 (83.4)	234.2 (103.2)	N/A	N/A	N/A	N/A
	Johnius belangerii	6.1 (2.5)	1.5 (0.4)	367.4 (107.5)	375.0 (109.5)	N/A	N/A	N/A	N/A
	Johnius heterolepis	4.6 (3.8)	1.0 (0.2)	407.5 (97.3)	413.2 (94.3)	N/A	N/A	N/A	N/A
	Pisodonophis cancrivorus	N/A	N/A	N/A	N/A	21.3 (36.9)	22.3 (38.5)	421.1 (89.2)	464.6 (43.5)
	Platycephalus indicus	0.0 (0.0)	49.2 (18.5)	1079.9 (378.7)	1129.2 (397.2)	55.3 (35.2)	47.0 (6.8)	219.5 (52.1)	321.8 (44.3)
	Scoliodon laticaudus	3.3 (5.6)	35.3 (8.8)	860.7 (168.4)	899.3 (177.3)	N/A	N/A	N/A	N/A
	Siganus canaliculatus	N/A	N/A	N/A	N/A	0.0 (0.0)	28.2 (15.2)	127.2 (77.0)	155.5 (92.1)
	Takifugu ocellatus	N/A	N/A	N/A	N/A	18.7 (13.2)	141.7 (74.8)	539.3 (276.8)	699.7 (364.6)
	Trypauchen vagina	N/A	N/A	N/A	N/A	0.0 (0.0)	36.0 (3.4)	101.8 (31.6)	137.8 (30.5)

Hong Kong and China in comparison to the rest of the world (IMO, 2008; Deng et al., 2015; HKLegCo, 2016). These results further demonstrated the necessity of monitoring of TPT in marine mammals of Pearl River Estuary, where major marine ports are located and hosts a few highly densely populated coastal cities like Hong Kong, Macau and Zhuhai with continuous TPT inputs into the marine environment associated with shipping activities, sewage discharge and surface runoff (Ho et al., 2016).

4.2. Inter-species, sex and maturity comparison of TPT contamination

In this study, Indo-Pacific humpback dolphin were found to be more contaminated than the finless porpoises. Such a considerable difference between the contamination status of the two species could be attributable to their diverging habitats, home range, and prey selections (Weisbrod et al., 2000). Indo-Pacific humpback dolphins generally inhabit the inner estuary of Pearl River located in the western waters of Hong Kong (equivalent to WI in this study), which favor estuarine prey such as the lionhead fish (*Collichthys lucidus*), croakers (*Johnius* spp.) and anchovies (*Thryssa* spp.) (Barros et al., 2004; Hung et al., 2006a). This area was found to be most contaminated by TPT and other organic compounds because it directly receives severely contaminated freshwater influx from surface runoffs and from the Pearl River (Wurl et al., 2006; Gui et al., 2014; Lai et al., 2016; Lam et al., 2016; Sham, 2019), discharges of partially treated wastewater from sewage treatment plants, and leachates from PT-coated ship hulls associated with heavy marine traffic in the region (Jefferson, 2000; HKEPD, 2009; Ho et al., 2016). Hence, Indo-Pacific humpback dolphins are likely to bioaccumulate PTs through uptake from the contaminated environment and food, as shown in the present study.

On the contrary, finless porpoises are known to have a greater home range from southwest (identified as WO in the currently study) to southeast waters of Hong Kong (Hung, 2018). The large home range of finless porpoises does not only allow them to gain greater exposure to cleaner environments, but also a wider range of food items of cephalopods and pelagic fishes, including squids (*Loligo* spp.), cuttlefish (*Sepiella* spp.), cardinalfish (*Apogon* spp.), and ponyfish (*Gazza minuta*) (Barros et al., 2002), that consequently led to a lower tissue concentration of TPT when compared to that of Indo-Pacific humpback dolphins.

For both marine mammal species, concentrations of PTs in muscle tissues of male individuals were generally higher than that in females in this study. Lower concentrations of TPT in females, in particular matured females, could be attributed to the transplacental transfer of TPT

Table 4
Statistical results of the regression analysis between the trophic level (TL) and log–concentrations of phenyltin (PT) residues (mono-, di-, and tri-phenyltin; MPT, DPT, and TPT) or total PT, in the marine organisms inhabiting Inner (WI) and Outer (WO) Estuary in western waters of Hong Kong, respectively. The calculated Trophic Magnification Factor (TMF) is also shown. ND denotes that the TMF was not determined due to data insufficiency. Bolded values are statistically significant at $p < 0.05$.

				Using 2.5 as enrichment factor			Using 3.4 as enrichment factor		
		R^2	*p*-value	Intercept	Slope^	TMF	Intercept	Slope^	TMF
WI	MPT	0.113	0.187	–2.003	0.51	ND	–2.373	0.70	ND
	DPT	0.301	**0.023**	–2.243	**0.78**	6.03	–2.806	**1.06**	11.48
	TPT	0.819	**< 0.001**	0.961	**0.39**	2.45	0.683	**0.53**	3.39
	PT	0.710	**< 0.001**	0.807	**0.46**	2.88	0.479	**0.62**	4.17
WO	MPT	0.038	0.407	0.101	0.32	ND	–0.131	0.44	ND
	DPT	0.145	0.098	0.437	0.36	ND	0.178	0.49	ND
	TPT	0.566	**< 0.001**	1.033	**0.40**	2.51	0.746	**0.54**	3.47
	PT	0.197	**0.050**	1.934	**0.23**	1.70	1.766	**0.32**	2.09

^ Significant slopes ($p < 0.05$) are bolded.

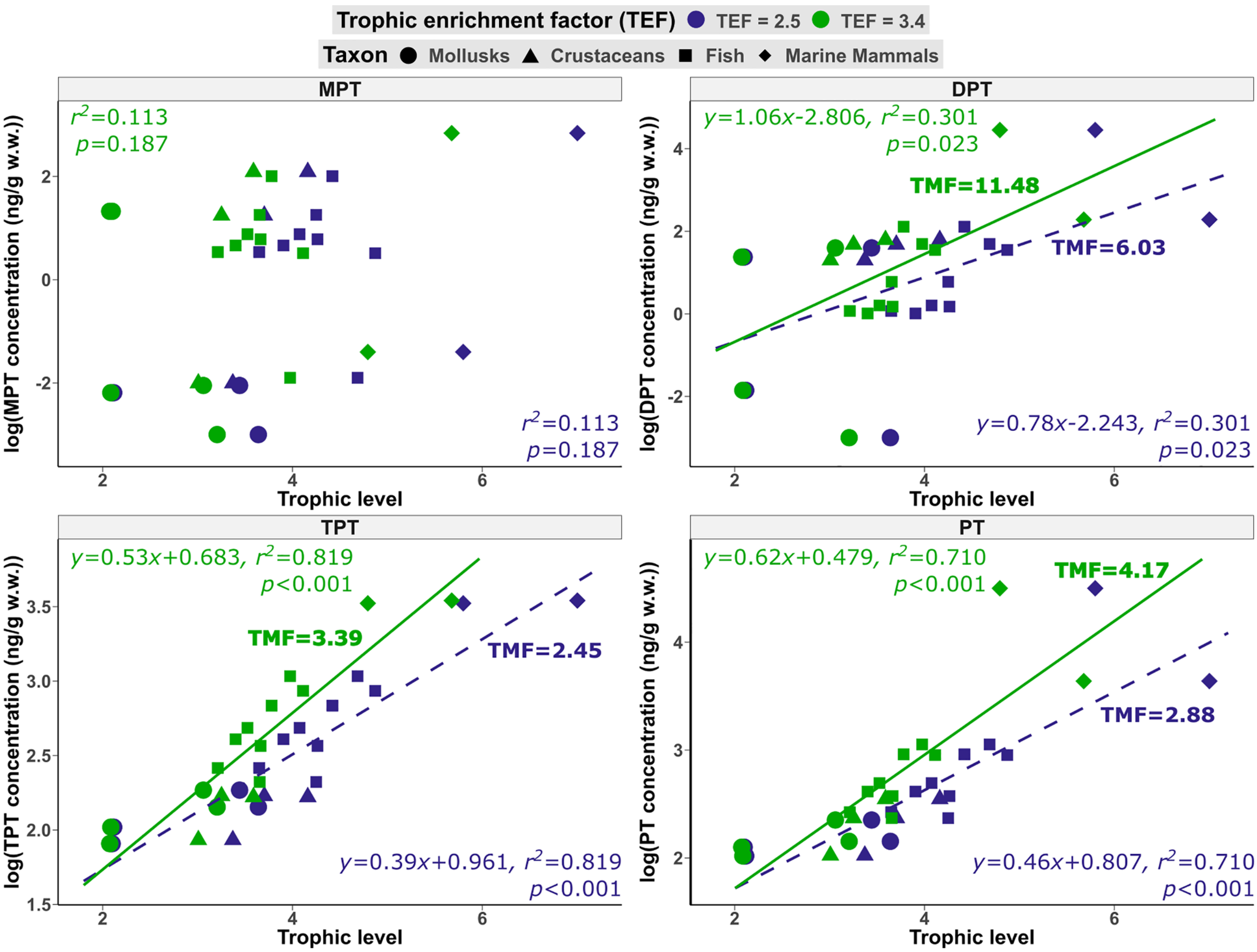

Fig. 3. Relationships between trophic level (TL) and log-transformed concentration of each PT residue (MPT, DPT, and TPT) and total PT in marine organisms obtained at the Inner Estuary (WI) in the western waters of Hong Kong. The results of regression analysis are provided for each sub-figure (purple dashed line: TEF = 2.5; green solid line: TEF = 3.4), and only significant relationship is plotted with a regression line.

from mother to fetus during gestation (Yang et al., 2007). Moreover, lactation has been suggested to be a more important factor for female mammals to eliminate persistent organic contaminants from their body (Hickie et al., 1999; Borgå et al., 2004), leading to elevated concentrations of PTs in neonate and calf individuals of Indo-Pacific humpback dolphins as shown in this study (total PTs concentrations: 1359.8–2795.5 ng/g w.w.; Table 1). Nevertheless, given the limited sample size in this study, a more in-depth investigation should be carried out to verify such hypotheses in future.

TPT concentrations in the finless porpoises was found to increase from juvenile, sub-adult, to adults. Likewise, higher TPT concentrations were generally observed in adults of the Indo-Pacific humpback dolphins than those in their neonates/calves. The increase in TPT concentration across maturity stages provided an evidence that long lifespans of marine mammals allow them to accumulate more xenobiotic contaminants through time (Bossart, 2011).

4.3. Structure of the selected food webs

The characterization of a food web, including the mapping of food webs and energy transfer across species, is essential because it provides a crucial foundation for evaluating the biomagnification of a compound within an ecosystem (Won et al., 2018). To understand the trophic relationships among the selected marine organisms, stable isotope analyses of carbon and nitrogen were used in the two studied sites to describe the trophodynamics of the two food webs in the current study. In particular, the carbon isotopes of organisms reflected the dietary sources of the various analyzed organisms, while the nitrogen isotopes described the relative trophic position of an organism within the food web (McCutchan et al., 2003).

The significant differences in $\delta^{13}C$ between organisms sampled from WI and WO reflected that marine organisms inhabiting the two food webs utilized varying dietary sources. Previous studies reported that sediment samples collected near the Pearl River mouth generally have more negative $\delta^{13}C$ values, while samples from the shelf of South China Sea displayed less negative $\delta^{13}C$ values (Hu et al., 2006a). Such a finding allows further differentiation of the dietary sources of organisms, in which more depleted $\delta^{13}C$ reflects higher assimilation of terrestrial-derived carbons, whilst more enriched $\delta^{13}C$ indicates more utilization of marine-derived carbons (Wai et al., 2011). Similar to the study conducted by Hu et al. (2006a), organisms in WI, including the Indo-Pacific humpback dolphins, were found to assimilate more depleted $\delta^{13}C$, indicating that these organisms favor estuary habitat and obtain food with carbons originated from terrestrial and estuarine sources (Hu et al., 2006a; Wai et al., 2011). Such a phenomenon also implies that these organisms have greater potential to be affected by the contaminants in the surface runoff and river discharges from the Pearl River Delta region. In contrast, the more enriched $\delta^{13}C$ detected in organisms from WO suggested that these organisms, including finless porpoises, utilize the more oceanic food sources (Chisholm et al., 1982;

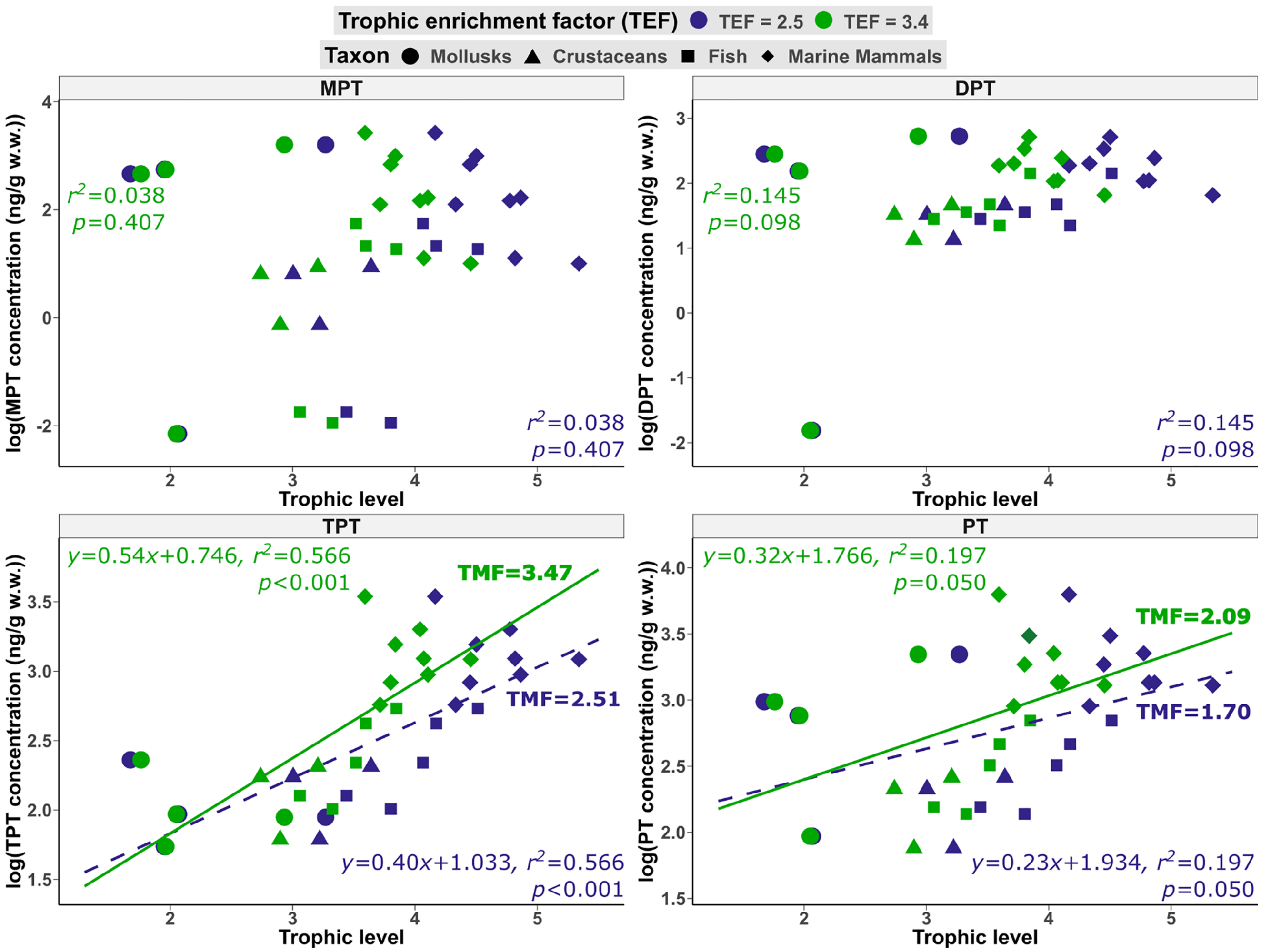

Fig. 4. Relationships between trophic level (TL) and log-transformed concentration of each PT residue (MPT, DPT, and TPT) and total PT in marine organisms obtained at the Outer Estuary (WO) of the western waters of Hong Kong. The results of regression analysis are provided for each sub-figure (purple dashed line: TEF = 2.5; green solid line: TEF = 3.4), and only significant relationship is plotted with a regression line.

Wai et al., 2011). These more oceanic waters are known to experience more oceanic dilution and thus receive less contamination in their food webs when compared with that of those adjacent to the inner Pearl River Estuary (Sham, 2019).

On top of the distinctive community isotopic niches presented in the current study, the community compositions of fish species were also found to be different between the two studied food webs. Fisheries survey conducted in the past decade have demonstrated a higher abundance and biomass in the WO than that in WI. However, the dominance of zoobenthivorous and piscivorous fishes in WI is generally greater than that in WO, while omnivorous fishes were found to occupy higher proportions in WO than in WI (Mak, 2017). This observed variation in fish communities of the two studied food webs is probably driven by spatial differences in environmental attributes (e.g., water depth, sediment profile and salinity gradient) and anthropogenic disturbances (e.g., water pollution, fishing-related activities and reclamation) (Wong, 2016). The availability of different fish species in WI and WO, in turn, controls the dietary preferences of the two local marine mammal species.

Furthermore, a mismatch in the trophic position was observed between the apex predators of the two studied food webs, in which the Indo-Pacific humpback dolphins in WI generally occupied higher trophic positions than that of finless porpoises in WO due to their differences in dietary preference (Barros et al., 2002; 2004). The relatively higher trophic position in Indo-Pacific humpback dolphins (TL: 4.79–7.00) also implies a greater magnification of PT contaminants in the dolphins than in the finless porpoise (TL: 3.59–5.34), thereby causing a higher tissue concentration of TPT in the dolphins as shown in this study (See Section 4.4).

4.4. Trophic magnification tendency of TPT and its degradation products

The evaluation of biomagnification of organic compounds is particularly critical because biomagnification can lead to a severely high chemical concentration in the top predators of a food web and trigger adverse effects on their fitness (Kelly et al., 2007). Since TMF has been suggested to be the best indicator of biomagnification potential (Conder et al., 2012), this study adopted and calculated TMF for determining whether TPT and its degradation products can be biomagnified in the food webs of the two resident marine mammal species.

The TMF values of TPT obtained from the two analyzed food webs (WI: 2.45–3.39; WO: 2.51–3.47) were consistently much larger than 1, confirming that TPT is biomagnified in higher trophic organisms through dietary uptake. The present results are in good agreement with the previously reported TMF values, which ranged from 1.1 to 5.3 (Supplementary Table 3), with an overall average of 3.5. Although numerous studies agreed that TPT can be biomagnified in lower trophic organisms of marine food webs (Hu et al., 2006b; Kono et al., 2008; Murai et al., 2008), contradictory findings were reported by Strand and Jacobsen (2005) that higher TPT concentrations were recorded in fishes compared to their hypothetical predators, i.e., fish-eating seabirds and harbor porpoises in Danish coastal water. Nonetheless, the study by Strand and Jacobsen (2005) covered a relatively wide area of ca. 200 km long from east to west, and ca. 160 km from north to south (Murai et al., 2008), and did not conduct any stomach content nor stable isotope analysis to verify the trophic position of each species and confirm predator-prey interactions of the food web. Such approach could lead to misinterpretation of the results obtained because the analyzed organisms did not necessarily belong to the same food web (Borgå et al., 2012). Moreover, Strand and Jacobsen (2005) investigated the biomagnification profile by comparing TPT concentrations in the whole organisms of invertebrates with that of the liver in organisms occupying high trophic levels. However, the concentrations obtained using this approach may not be comparable because high trophic organisms might not consume the whole prey when food is abundant (Nelson et al., 1983), and concentrations in the liver merely reflected the sink of xenobiotic chemicals in the organisms (Gray, 2002). More importantly, previous studies have discovered that the concentration of TPT in liver could not serve as an accurate predictor of the total TPT concentration in the whole organism in fishes (Franklin, 2016; Sham, 2019), and therefore such an inappropriate comparison could lead to a faulty conclusion.

Further to the trophic magnification potential of TPT, the current study also looked at TMF values of other PT residues, including DPT, MPT, and total PT. Unlike TPT, the trophic magnification of DPT and MPT exhibited a site-specific profile; for instance, DPT was magnified through trophic cascade in WI but not in WO.

4.5. Environmental implications for future management measures

Traditionally, biomagnification tendency of a compound is inferred by the hydrophobicity/lipophilicity of the target compound, which chemicals with an octanol-water partition coefficient, log $K_{OW} \geq 5$ are commonly considered to be biomagnifiable in the aquatic food web, whereas chemicals with log $K_{OW} < 5$ demonstrate the otherwise (Borgå et al., 2012). However, recent studies have suggested that log K_{OW} cannot serve as a universal model for identifying biomagnification potential of all synthetic organic substances (Kelly et al., 2007). In particular, studies suggested that amphiphilic substances, such as ionogenic compounds, have the affinity to both water and lipids, and therefore the prediction of biomagnification potential through the use of log K_{OW} would not reflect the real-life biomagnification scenario (Ehrlich et al., 2011; Mcdougall, 2016). The results of the current study demonstrated that, TPT, which has a log K_{OW} ranged between 3.0 and 4.0 (WHO, 1999), has a TMF that comparable with the other highly bioaccumulative lipophilic organic contaminants that have log K_{OW} values > 5, including polychlorinated biphenyl (PCBs), polybrominated diphenyl ethers (PBDEs), Dichlorodiph enyltrichloroethane (DDT), perfluorinated compounds (PFCs), and other organophosphate flame retardants (OPFRs) (Mizukawa et al., 2013; Walters et al., 2016). Similar to TPT compounds, some persistent organic compounds such as trichlorinated benzenes, lindane, alpha- and beta-HCH have been found to manifest a significant trophic magnification in marine food webs despite having a log $K_{OW} < 5$ (Hop et al., 2002; Hoekstra et al., 2003; Mackintosh et al., 2004). Such findings demonstrated that TMF is a more convincing indicator of biomagnification in the marine food web regardless to the log K_{OW} value of the contaminant of concern.

The results of this study provide solid evidence to encourage environmental authorities to prudently consider the bioaccumulation and biomagnification potentials of ionizable compounds like TPT and DPT when assessing and prioritizing their risks to the marine ecosystem and human health. Remarkably, more than 37% of the "compounds of concerned" in the United States Environmental Protection Agency's BCFBAF program are considered to be ionizable compounds (Costanza et al., 2012). More importantly, air-respiring organisms are suggested to have high rates of accumulation but low rates of elimination of ionizable compounds (Kelly et al., 2007), advocating that the accurate determination of the bioaccumulation and biomagnification potentials of these compounds is crucial to the protection of the marine environment and human health.

5. Conclusions

The two resident marine mammals, namely Indo-Pacific humpback dolphin and finless porpoise, were adopted as indicator species to access the contamination status of PTs, including MPT, DPT, and TPT in Hong Kong's western waters. With the high concentrations of PTs detected in these two resident marine mammals, the Pearl River Delta region remains a pollution hotspot of PTs, even though the International Maritime Organization (IMO) has implemented a global ban of application of PT compounds in antifouling coatings on seagoing vessels since September 2008 (IMO, 2008). Evidently, TPT was consistently biomagnified through the marine food web and exhibited the highest concentrations in the top predators, i.e., the two resident marine mammals. Hence, the bioaccumulation and biomagnification potentials of TPT and its degradant (DPT) should be considered when assessing the ecological risks of these compounds. The bioaccumulation and biomagnification of TPT and DPT can be further inferred in a further study that comprehensively evaluates human health risks associated with the exposure and bioaccumulation of these compounds via dietary intake.

CRediT authorship contribution statement

Ronia C.T. Sham: Conceptualization, Methodology, Investigation, Formal analysis, Writing - original draft, Writing - review & editing. **Lily S.R. Tao:** Investigation, Formal analysis, Writing - review & editing. **Yanny K. Y. Mak:** Investigation, Formal analysis. **Jason K. C. Yau:** Investigation, Formal analysis. **T. C. Wai:** Methodology, Investigation, Writing - review & editing. **Kevin K. Y. Ho:** Methodology, Writing - review & editing. **G. J. Zhou:** Formal analysis, Writing - review & editing. **Y.-Y. Li:** Methodology, Investigation, Supervision, Writing - original draft. **X.-H. Wang:** Conceptualization, Funding acquisition, Methodology, Resources, Supervision, Writing - original draft, Writing - review & editing. **Kenneth M. Y. Leung:** Conceptualization, Formal analysis, Funding acquisition, Methodology, Project administration, Resources, Supervision, Writing - original draft, Writing - review & editing.

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Appendix A. Supplementary material

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