Risk Assessment Studies

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Organotin Compounds in Aquatic Products Available at Local Markets

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Centre for Food Safety

Food and Environmental Hygiene Department

The Government of the Hong Kong Special Administrative Region

Correspondence:

Risk Assessment Section

Centre for Food Safety

Food and Environmental Hygiene Department

43/F, Queensway Government Offices,

66 Queensway, Hong Kong.

Email: enquiries@fehd.gov.hk

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EXECUTIVE SUMMARY

Organotin compounds (OTC) are a large class of compounds composed of tin (Sn) directly bound to different organic groups and they differ in their properties and applications. Among different types of OTC, tri-substituted compounds, in particular tributyltin (TBT) and triphenyltin (TPT), have been widely used as biocides in wood preservatives, algicides and molluscicides in antifouling products on ship as well as pesticides for agricultural purposes. Mono- and di-substituted OTC like monomethyltin (MMT), dimethyltin (DMT), dibutyltin (DBT), mono-n-octyltin (MOT) and di-n-octyltin (DOT) are generally used in mixtures in various amounts as polyvinyl chlorides (PVC) stabilisers.

2. TBT and TPT can cause masculinisation (imposex) in female snails at low concentrations (1 ng/L in water), suggesting that these compounds are endocrine disruptors. They are toxic to aquatic organisms and tend to bioaccumulate through the food chain, in particular in fish and other seafood, thus consumption of contaminated aquatic products may pose a potential health risk to humans. A group tolerable daily intake (TDI) of 0.25 mcg/kg bw/day (as TBTO), equivalent to 0.1 mcg/kg bw/day (when expressed as Tin (Sn)) for OTC including TBT, TPT, DBT and DOT was established in 2004 by the European Food Safety Authority (EFSA).

3. The objectives of this study are (1) to determine the levels of four OTC (i.e TBT, DBT, TPT and DOT) in aquatic products available in

local markets; (2) to estimate the dietary exposure to the four OTC of Hong Kong adult population; and (3) to assess the associated health risk.

4. A total of 341 samples of fish, molluscs and crustaceans were collected and tested for four OTC. Results showed that 205 samples (60%) were detected with one or more OTC, with TPT most commonly detected (53% of samples).

5. The total OTC levels in the samples ranged from below detection limit to 490 mcg/kg, expressed as Sn. As for the mean total OTC levels in different food groups, fish contained the highest level at 24 mcg/kg as Sn, followed by molluscs (15 mcg/kg as Sn) and crustaceans (14 mcg/kg as Sn).

6. The dietary exposures of both the average and high consumers (90^{th} percentile) of the local adult population to the total OTC were estimated to be 0.020 and 0.057 mcg/kg bw/day, respectively. Comparing the dietary exposures with the health based guidance value (HBGV), they accounted for 20% and 57% respectively of the group TDI established by EFSA in 2004. The food group fish was the major contributor to the OTC dietary exposure of the local population (88%).

Conclusions and Recommendations

7. The dietary exposures of both the average and high consumers of the local adult population to the OTC were below the HBGV, indicating

that adverse health outcome due to OTC exposure of the Hong Kong adult population from aquatic products commonly available at local markets was unlikely.

8. The public is advised to maintain a balanced and varied diet which included a wide variety of meat, vegetables and fruits so as to avoid excessive exposure to any contaminants from a small range of food items. As fish contain many essential nutrients, such as omega-3 fatty acids and high quality proteins, it is therefore advisable to consume a variety of fish and not to overindulge in only a few species.

9. Reduction of OTC in aquatic products relies mainly on the control of their use and release in order to safeguard the marine ecosystem and human health. With the increasing number of places to restrict the use of OTC in antifouling paints for ships and pesticides, OTC in the environment are expected to be in a decreasing trend.

Risk Assessment Studies –

Organotin Compounds in Aquatic Products Available at Local Markets

OBJECTIVES

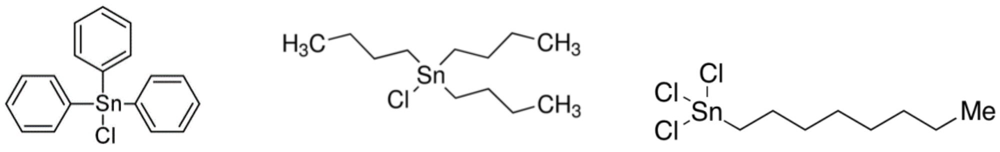
This study aims (i) to determine the levels of four organotin compounds (OTC), namely TBT, DBT, TPT and DOT in aquatic products available in local markets; (ii) to estimate the dietary exposure to the four OTC of Hong Kong adult populations; and (iii) to assess the associated health risk by comparing the estimated dietary exposure with the health-based guidance value (HBGV).

BACKGROUND

2. Organotin compounds (OTC) are a large class of compounds composed of tin directly bound to different organic groups and they differ in their properties and applications. Where tin (Sn) is a natural element in the earth's crust which can be used to line cans for food, beverages and aerosols.

3. OTC can be divided into four groups with the general formulas R_4Sn, R_3SnX, R_2SnX2, and $RSnX_3$, where R is usually an organic group and X is an anion (e.g. chloride, fluoride, oxide or hydroxide).

Figure 1. Structure of some organotin compounds



(a) Triphenyltin chloride; (b) Tributyltin chloride (c) Mono-n-octyltin trichloride

4. OTC have many applications. Tri-substituted OTC, in particular tributyltin (TBT) and triphenyltin (TPT), have been extensively used as biocides to preserve wood, algicides and molluscicides to prevent fouling of boats and hulls of ship as well as pesticides for the agricultural applications since the 1960s. Mono- and di-substituted OTC like monomethyltin (MMT), dimethyltin (DMT), dibutyltin (DBT), mono-n-octyltin (MOT) and di-n-octyltin (DOT) are generally used in mixtures in various amounts as polyvinyl chlorides (PVC) stabilisers for food contact materials[1,2]. Therefore they are present in antifouling paints, PVC food packaging materials, agrochemicals and many other consumer products. Through leaching from the antifouling paint used in boat and ship and other marine equipment, particular in area of high shipping activity such as ports, marinas and shipyards, from the PVC products disposed into sanitary landfills, as well as from the runoff of agricultural fields, these compounds have been introduced into our estuarine and marine ecosystems[3,4].

Sources of Dietary Exposures

5. As OTC are relatively persistent in the environment and have a tendency to bioaccumlate in the marine ecosystem through the food chain, the general public is exposed to OTC mostly through intake of fish and other seafood [1]. Other possible sources of dietary exposure could be from pesticides, additives used in plastics and other food contact materials. According to literature, the levels of OTC detected in foods other than seafood such as cereals, meat, fruits and vegetables were relatively low[5].

Toxicity

Kinetics and metabolism[1,6]

6. OTC hydrolyse in aqueous media. For example, TPT compounds containing an anionic group such as chloride (TPTCl) or acetate (TPTA) can hydrolyse to its hydroxide (TPTH) at ambient temperatures in the pH range of 3 - 8. Accordingly, hydrolysis of OTC can occur in food including aquatic organisms. Dietary studies with e.g. TPTA (or TPTCl) would therefore predict the oral toxicity of triphenyltin hydroxide (TPTH). After oral uptake, various OTC may be converted in part to their chlorides (OTCl) which are absorbed in the gastro-intestinal tract. OTC possess both lipophilic and ionic properties – the former favours their accumulation in lipids while the latter enables OTC to bind to proteins and glutathione. In rats, TBTO was absorbed

incompletely and slowly from the gastrointestinal tract and the principal route of excretion for OTC is via faeces with smaller percentage in urine with their metabolites if metabolism has taken place.

Acute toxicity

7. Animal studies showed OTC have medium acute toxicity[1]. Effects of acute exposure following oral exposure to TPTH may include anorexia, tremor, and diarrhea, followed by drowsiness and ataxia[7]. After dermal exposure to TPTH, skin irritation was observed. With five applications of 100 mg/kg to five rats respectively, it caused death to one animal and local sign of irritation. With a single dermal application of 12.5 mg TPTA/kg to three rabbits, it was lethal for two rabbits and caused black coloration of the skin was scarcely pilous sites of the body[8]. With an exposure of 100mg TBTO/kg body weight by oral gavage, a transient increase in adrenal weight was observed shortly after exposure and a transient effect on thyroid follicles was observed. Also signs of irritation (for example, nose discharge and lung edema) and enteritis were observed after a single 4-hour exposure of rats to aerosol of TBTO[9].

Chronic toxicity[1]

8. TBT, DBT, TPT and DOT affect the immune system. The lowest no-observed-adverse-effect level (NOAEL) detected in the toxicity tests for TPTH was 0.1 mg /kg bw/day (as TPTH[6]) for maternal toxicity in a rabbit gavage study, based on decreased food consumption

and body weight gain at 0.3 mg/kg body weight per day, whereas the reported NOAEL for TBTO in immunotoxicity test from chronic feeding studies in rats was 0.025 mg/kg bw/day (as TBTO).

Carcinogenicity and genotoxicity

9. TPT and TBTO have not been assessed for carcinogenicity by the International Agency for Research on Cancer (IARC) but some data showed that TPT are co-lastogenic (i.e. combined effect in inducing disruption or breakages of chromosomes). The United States Environmental Protection Agency (U.S. EPA) has evaluated TPTH as category B2 ("probable human carcinogen")[10], and assigned TBTO as "cannot be determined"[11]. TBT and TPT do not present a genotoxic hazard[1].

Endocrine disrupting effects

10. According to toxicology evaluations on organotins by the European Food Safety Authority (EFSA) and the World Health Organization (WHO)[6,7], both TBT and TPT exert deleterious effects on aquatic organisms even at very low concentrations (1 ng/L). The compounds could lead to masculinization (or "imposex") in female snails at low concentrations (1 ng/L in water), a phenomenon in which female gastropods develop male sex organs, thus demonstrating the ability of these compounds to behave as endocrine disruptors for some aquatic organisms. Imposex gives rise to reproductive failure and as a

consequence to population decline. There are also reports showing reproductive and developmental toxicity in rodents at relatively low doses (around 1 mg/kg bw/day) which further supports the presence of endocrine activity.

Health-Based Guidance Value

11. The Joint FAO/WHO Expert Committee on Food Additives (JECFA) has not established any HBGV for OTC. According to the EFSA, the most toxic OTC are TBT, DBT and TPT and they are primarily found in fish and fishery products. In particular, TBT and TPT are highly toxic to aquatic organism and show a complex toxicity profile in rodent. Furthermore, the EFSA considered that DOT exerts similar immunotoxicity through a similar mode of action when compared with TBT, DBT and TPT. In 2004, the EFSA identified a no observed adverse effect level (NOAEL) for immunotoxicity of 0.025 mg/kg bw/day (as TBTO) from chronic feeding studies. Because TPT, TBT, DBT and DOT exert their immunotoxic effects by similar mode of action and potency, the EFSA considered it reasonable to establish a group tolerable daily intake (TDI). By applying a safety factor of 100, a group TDI of 0.25 mcg/kg bw (as TBTO), equivalent to 0.1 mcg/kg bw /day (when expressed as Tin (Sn)) for TPT, TBT, DBT and DOT compounds was established.

Regulatory Control on the Use of Organotin Compounds

International situation

12. Owing to the recognised potential adverse effects of OTC on both humans and aquatic ecosystems, many countries have banned organotin-based antifouling paints[12,13]. For instance, the European Commission Parliament adopted a regulation (No. 78/2003) which banned the use of all organotin-based antifouling paints on all vessels of European member countries since 2003[14]. Japan and New Zealand have banned the use of TBT antifouling paints on all vessels[12]. The International Maritime Organization (IMO) adopted the "International Convention on the Control of Harmful Anti-Fouling Systems on Ships, 2001" (the Convention) to prohibit the use of OTC in anti-fouling paints used on ships and the Convention entered into force internationally in 2008[15]. The Convention recommends the prohibition of application of OTC in any antifouling system worldwide so as to protect the marine environment. In June 2011, Mainland China has implemented the Convention and banned the use of organotins on vessels entering Mainland China[16].

Local situation

13. In Hong Kong, all antifouling paints with biocides are considered as pesticides and are subject to regulatory control under the Pesticides Ordinance (Cap.133) administered by the Agriculture,

Fisheries and Conservation Department (AFCD)[17]. The regulation stipulates that only antifouling paints which are registered pesticides may be imported, supplied and sold by licensed pesticide traders for use in Hong Kong. Antifouling paints which are not registered as mentioned above are controlled under a permit system where no person may import, supply, sell, possess and use any unregistered antifouling paints unless he/she is in possession of a valid pesticide permit issued by the AFCD.

14. Furthermore, the Merchant Shipping (Control of Harmful Anti-Fouling Systems on Ships) Regulation (Cap. 413N) has come into operation since on 1 January 2017 to implement the International Convention on the Control of Harmful Anti-Fouling Systems on Ships 2001. The Regulation is applicable to all Hong Kong registered ships wherever they are, foreign ships within Hong Kong waters, as well as local vessels as defined in the Merchant Shipping (Local Vessels) Ordinance (Cap. 548)[18]. The Regulation prohibits the use of organotin compounds that act as biocides in the anti-fouling systems of ships for the protection of the marine environment.

15. The AFCD has stopped issuing permits for the import, supply and sales of anti-fouling paints containing organotin compounds (OTC) since 2006[19]. Currently, no OTC-containing antifouling paint are registered under Cap. 133[20]. Like many places including European Union[21] and Australia[22], no organotin pesticides are registered in Hong Kong[23,24] for agricultural purposes.

Regulation of Organotin Compounds in Food

16. The Codex Alimentarius Commission (Codex) has not established any standard for OTC in food. In some places such as the United States, European Union, Singapore, Mainland China and Hong Kong, have set maximum residue limits (MRLs) for certain OTC in food when they are used as pesticides.

SCOPE OF STUDY

17. Aquatic products, including different types of fish, crustaceans and molluscs were collected from various retail markets for the analysis of four OTC (i.e TBT, DBT, TPT and DOT).

METHODOLOGY AND LABORATORY ANALYSIS

Methodology

18. A total of 341 samples of aquatic products were collected from the local markets between October 2017 and June 2018 in Hong Kong. For the purpose of this study, these samples were classified into three groups, namely fish, crustaceans, and molluscs. Aquatic products which are commonly consumed by Hong Kong population and those which have potentially higher risk, such as predatory fishes (e.g. tuna, mackerel and cod) and demersal fish (e.g sole and turbot) were selected

for testing (Table 1). A complete list of the types of food samples collected could be found in Appendix I.

Table 1. Samples of aquatic products

Food group (including their products)	**Number of Samples**
Fish (e.g crass carp, salmon, tuna, sea bass, pomfret, sole, mangrove snapper, etc.)	201
Crustaceans (e.g crab, shrimp, lobster)	31
Molluscs (scallop, mussel, squid, cuttlefish, sea cucumber)	109
Total	**341**

19. The sum of the four OTC levels in the aquatic products was combined with food consumption information captured from the Hong Kong Population-based Food Consumption Survey (2005-2007)[25], where food consumption data were collected from respondents on two non-consecutive days of 24-hour dietary intake, to obtain the dietary exposures to OTC of local adult population. The mean and 90th percentile exposure levels were used to represent the dietary exposures of the average and high consumers of the local population respectively.[i] The selection of the 90th percentile to represent high consumer is in line

[i] The 95thpercentile exposure level has been used by the CFS to represent the dietary exposure level of the high consumer of the local population (consumers only). However, international and national food/health authorities (e.g. FAO/WHO, US FDA and FSANZ) have expressed that, based on 1-, 2- or 3-day consumption survey data, the estimation of exposure for high consumers at 95th percentile may be over-estimated.

with the international practices where, based on one to three days of food consumption data, a high consumer's chronic dietary exposure is best represented by the 90th percentile exposure[26,27,28]. The estimation of dietary exposure was performed with the aid of an in-house developed web-based computer system, Exposure Assessment System (EASY).

20. The estimated exposure levels were then compared with the group TDI of 0.1 mcg/kg bw /day (expressed as Tin (Sn)) in order to assess the associated health risk.

Laboratory Analysis

21. Laboratory analysis of OTC was conducted by the Food Research Laboratory (FRL) of CFS. In this project, edible portion of 341 samples of aquatic products have been tested for four OTC including TPT, TBT, DBT and DOT. The collected samples were analysed as consumed. Certain samples were prepared by steaming, while some other samples like sashimi were analysed as purchased without further cooking.

22. The OTC levels in aquatic products were analysed by gas chromatography – inductively coupled plasma mass spectroscopy. Internal standard compound, tripentyltin was spiked quantitatively into a measured amount of sample, which was then extracted by vertical shaking with acidified methanol. The sample extract was filtered through Florisil. The sample solution was then extracted to dichloromethane (DCM). The DCM layer was purified with a Florisil

solid phase extraction cartridge. The OTC in eluent was ethylated before instrument analysis. Identification was confirmed by comparing the relative retention time with those of the standards. The limits of detection (LODs) and the limits of quantification (LOQs) of the organotin compounds were 0.25 and 1.0 mcg Sn kg^{-1} respectively.

Treatment of Analytical Values Below the LOD

23. In this study, data were treated with the lower bound (LB) and upper bound (UB) approach. That is, at the LB, results below the LOD were replaced by zero whilst at the UB, results below the LOD were replaced by the value reported as the LOD. This approach compares the two extreme scenarios, based on the consideration that the true value for results less than LOD may actually be any value between zero and the achieved LOD. The LB scenario assumes that the chemical is absent; therefore, to results reported as <LOD a value of zero is assigned. The UB scenario assumes that the chemical is present at the level of the LOD; thus, to results reported as <LOD a value of the corresponding LOD is assigned.

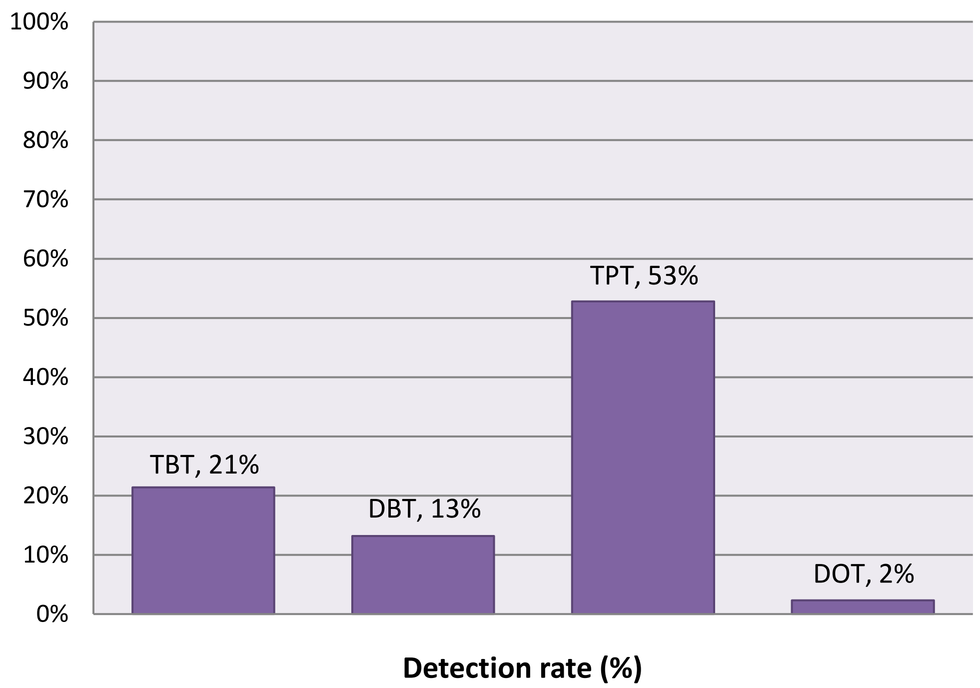
RESULTS AND DISCUSSION

24. Of the total of 341 samples of fish, crustaceans, and molluscs were collected from the local market for the analysis of OTC (namely TBT, DBT, TPT and DOT), 205 samples (60%) have at least one OTC detected at quantified levels. In other words, 136 samples (40%) were

not detected with any OTC. Levels of OTC detected in the samples could be found in Appendix I - IV.

25. Among the four OTC examined, TPT was the most commonly detected OTC and was found in 180 samples (53%), followed by TBT in 73 samples (21%) and DBT in 45 samples (13 %). DOT was least detected, in only 8 samples (2%). (Figure 2)

Figure 2. Detection rate of organotin compounds in food samples (in %)



26. The maximum levels of individual OTC detected in the aquatic samples: TPT at 1400 mcg/kg (480 mcg/kg as Sn), TBT at 60 mcg/kg (24 mcg/kg as Sn), DBT at 8.8 mcg/kg (4.5 mcg/kg as Sn), and DOT at 2.6 mcg/kg (0.89 mcg/kg as Sn) at LB estimation. (Table 2)

Table 2. Range of OTC levels* (LB and [UB]) (in mcg/kg) detected in the samples

TBT (LB)	ND** – 60 (ND** – 24) as Sn
TBT (UB)	[0.61 – 60] [0.25– 24] as Sn
DBT (LB)	ND** – 8.8 (ND** – 4.5)as Sn
DBT (UB)	[0.49 – 8.8] [0.25– 4.5] as Sn
DOT (LB)	ND** – 2.6 (ND** – 0.89)as Sn
DOT (UB)	[0.73 – 2.6] [0.25 – 0.89] as Sn
TPT (LB)	ND** – 1400 (ND** – 480) as Sn
TPT (UB)	[0.74 – 1400] [0.25 – 480] as Sn

*Rounded to 2 significant figures

**ND: Not detected

LODs of OTC = 0.25 mcg/kg as expressed in Sn, whereas LODs are ^0.61 mcg/kg for TBT, ^0.49 mcg/kg for DBT, ^0.7275 mcg/kg for DOT and ^0.7375 mcg/kg for TPT when expressed in their molecular ion forms

^LODs for TBT, DBT, DOT and TPT were calculated based on the given conversion factor for organotin compounds from Sn to Molecular ion (i.e. DBT/Sn = 1.96; TBT/Sn = 2.44; DOT/Sn = 2.91; TPhT/Sn = 2.95)

27. As regards the levels of four OTC in different aquatic product groups, the levels of TPT were found to be consistently the highest in all groups. The UB and LB mean concentration of TPT in fish were 69

mcg/kg (23 mcg/kg as Sn) and in molluscs were 43 mcg/kg (15 mcg/kg as Sn). The mean concentration of TPT in crustaceans was 41 mcg/kg at LB (14 mcg/kg as Sn) and 42 mcg/kg at UB (14 mcg/kg as Sn). Whereas all LB and UB mean levels of TBT, DBT and DOT (except UB mean level of TBT in fish) in the three aquatic product groups were below 1 mcg/kg (Table 3).

Table 3. Mean organotin compounds levels* (in mcg/kg) detected in the three groups of samples

group	fish	crustacean	molluscs
	Mean Levels (in mcg/kg)		
TBT (LB)	0.77 (0.32 as Sn)	0.38 (0.16 as Sn)	0.46 (0.19 as Sn)
TBT (UB)	1.27 (0.52 as Sn)	0.85 (0.35 as Sn)	0.90 (0.37 as Sn)
DBT (LB)	0.07 (0.04 as Sn)	0.32 (0.16 as Sn)	0.60 (0.31 as Sn)
DBT (UB)	0.54 (0.28 as Sn)	0.76 (0.39 as Sn)	0.94 (0.48 as Sn)
DOT (LB)	0.03 (0.01 as Sn)	0.0 (0.0 as Sn)	0.06 (0.02 as Sn)
DOT (UB)	0.75 (0.26 as Sn)	0.73 (0.25 as Sn)	0.76 (0.26 as Sn)
TPT (LB)	69 (23 as Sn)	41 (14 as Sn)	43 (15 as Sn)
TPT (UB)	69 (23 as Sn)	42 (14 as Sn)	43 (15 as Sn)

*Rounded to 2 significant figures

28. By summation of the four OTC concentration detected in as Sn form, the total OTC levels (LB) in all 341 samples ranged from below LOD to 490 mcg/kg as Sn. For the mean concentration of total OTC levels (LB) in different food groups, fish contained the highest level at 24 mcg/kg as Sn, followed by molluscs (15 mcg/kg as Sn) and crustaceans (14 mcg/kg as Sn). For details please refer to Appendix V.

Dietary Exposure to Organotin Compounds

29. The study results revealed that the dietary exposures to the total OTC from aquatic products of the local population (consumers only) were 0.020-0.021 mcg/kg bw/day (20-21% of the TDI) and 0.0574-0.0588 mcg/kg bw/day (57-59% of the TDI) for average and high consumers respectively at LB and UB estimation (body weight 61.13 kg, number of consumers: weighted = 3 743 164 and unweighted = 3 507, group TDI= 0.1 mcg/kg bw/day as Sn from EFSA 2004).

30. As the dietary exposures to the total OTC of both the average and high consumers of the adult population were below the HBGV, adverse health outcome due to OTC exposure of the Hong Kong adult population from aquatic products commonly available at local markets was unlikely.

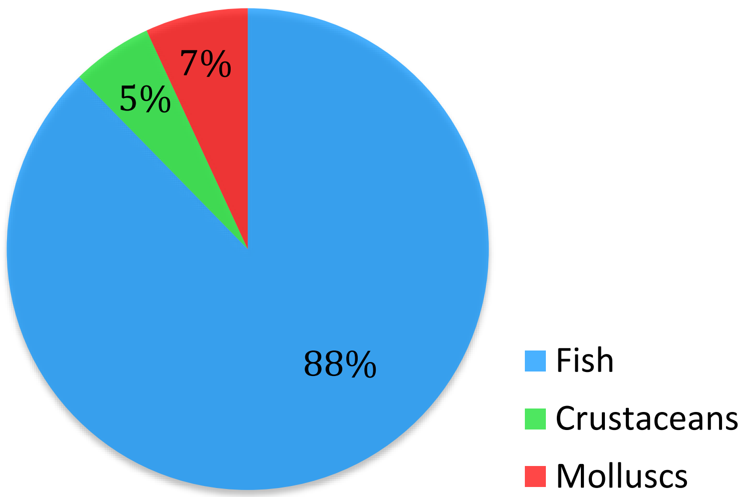
31. Among the three groups of aquatic product, the food group fish was found to be the major contributor (0.018 mcg/kg bw/day, 88% of the dietary exposure) which was related to the higher consumption amounts and relatively higher OTC levels detected in fish. The contribution of different food groups to the dietary exposure to the total OTC for an average consumers is summarised in Table 4 and Figure 3.

Table 4. Dietary exposure to organotin compounds for average consumers and percentage contribution of different groups (LB estimation)

Food groups	Dietary exposure* (mcg/kg bw/day)	% contribution
Fish	0.018	88
Crustacean	0.001	5
Molluscs	0.001	7
Total	**0.020**	**100**

* calculated and expressed in (mcg/kg bw/day) as Sn

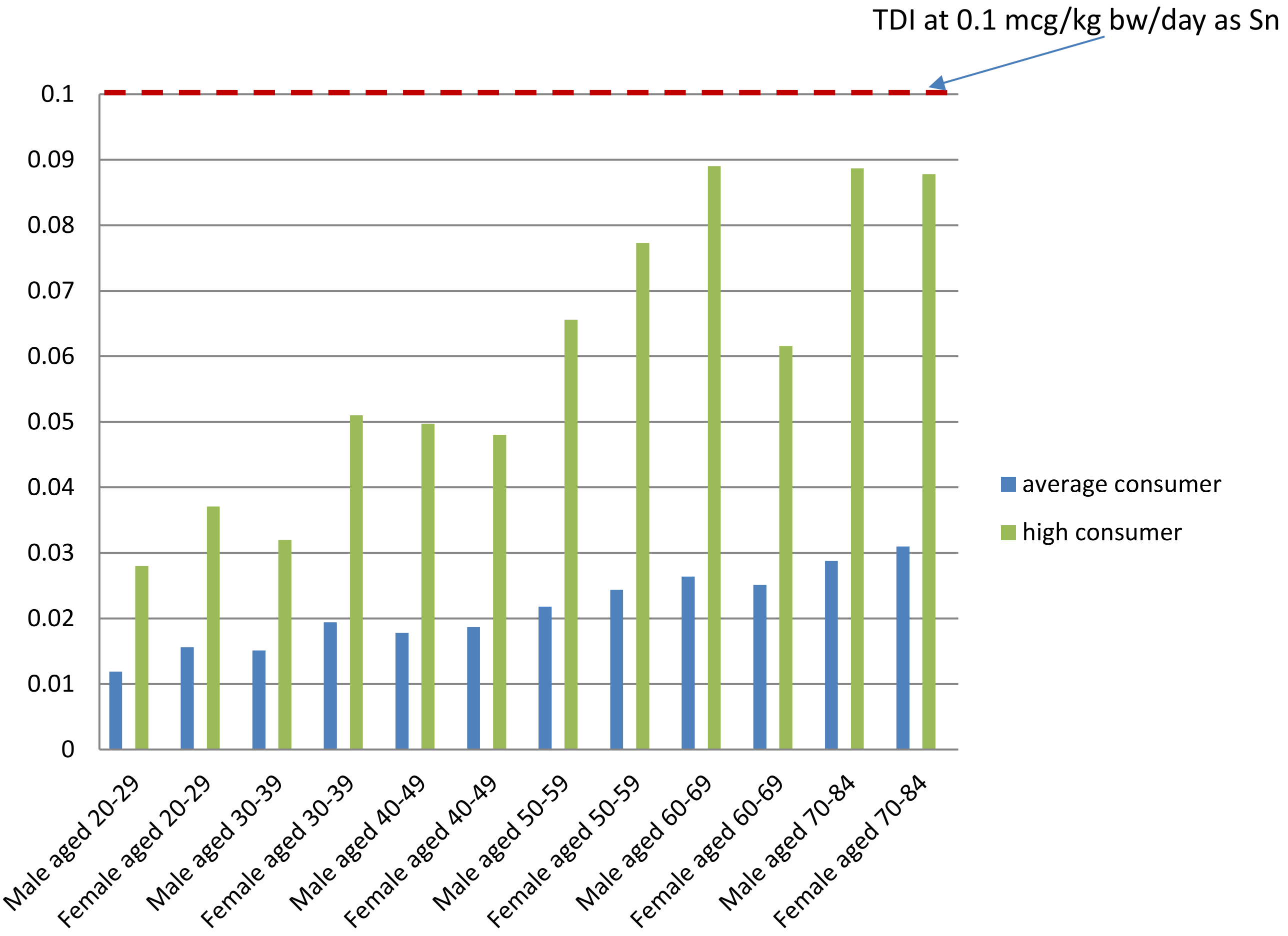
Figure 3. Relative contribution of each food group to dietary exposure to organotin compounds among average consumers in local adult population



32. Further analyses of dietary exposure of individual age-gender population subgroups were shown in Figure 4. The results showed that the dietary exposures to total OTC for all age-gender population

subgroups of average and high consumers were below the HGBV, meaning that the dietary exposure to OTC due to consumption of aquatic products did not indicate a heath concern for all age-gender subgroups.

Figure 4. Dietary exposure to organotin compounds by age-gender group (average and high consumers of the population)



International Situation

33. The prevalence as well as the concentration of different types of OTC vary in different studies, indicating that the sources of contamination might be different in various places. In our study, TPT

is the dominant type of OTC detected in aquatic products. This observation is the same as findings in another local study[29] and in Taiwan[30]. However, TBT is more commonly found than TPT in fish and other seafood in European countries[1]. The mean concentration (mcg/kg) of OTC in aquatic products in the current study and other European countries are summarised in Table 5. The difference in occurrence of OTC distribution in marine environment could depend on different factors, such as distance from OTC contamination sources and shipping activities. The OTC levels in general are higher in coastal areas such as harbours, estuaries or marina due to intense shipping activities[31].

34. The dietary exposure expressed as the percentage of HBGV in this study was compared to those obtained from other places and are summarised in Table 6. It was found that for average consumers, the percentage of HBGV in our study is lower than that of Norway but higher than that of Germany, France, Ireland, Finland and Korea. This observation could be related to the difference in consumption amount of aquatic products. According to the EFSA study in 2004[1], the average intake of fish and seafood products for people in Norway was 80 g per day per person whereas people in Hong Kong consumed a comparatively lower amount at around 68 g per day per person. However, it should be noted that direct comparison of data has to be done with caution due to the differences in time when the studies were carried out, research methodology, sampling strategies, approaches of capturing consumption

data, analytical methods, approaches of treating analytical results not detected limits, etc.

35. The Convention from IMO has entered into force since September 17 2008, to prohibit the usage of OTC in antifouling paints and with the increasing number of countries to restrict the use of OTC in antifouling paints for ships and pesticides, OTC in the environment are expected to be in a decreasing trend. This phenomenon has been reported in some studies conducted in estuarine areas of Japan, South Korea and some European countries[32,33,34].

Table 5. Concentration of OTC (mcg/kg) as Sn in aquatic products

Places	**Mean concentration of OTC^ (mcg/kg) as Sn**		
	TBT	**TPT**	**DBT**
Hong Kong 2018 (Current study)	**0.26**	**20**	**0.13**
European countries participating in the SCOOP task (EFSA 2004)[1]	**12#**	**5.8#**	**8.6#**

^ Concentration of DOT was not shown as the EU SCOOP report contains very few data on DOT, which were always below the limit of detection.

International mean concentration of TBT, DBT and TPT of fish and fishery products from European countries participating in the SCOOP task (EFSA 2004)

Table 6. Percentage of health-based guidance value for average consumer in different places

Places	% of HBGV^,ξ Average consumer
Hong Kong 2018 (Current study)	20
Norway 2004[1]	33#
Germany 2004[1]	12#
France 2004[1]	17#
Ireland 2004[1]	9.6#
Finland 2010[35]	1.3
Korea 2012[36]	6.9^^

^ % of HBGV was estimated by comparing the dietary exposures to OTC being studied in the countries to the HGBV for average consumers.

ξHBGV for group OTC (TDI) is defined as 0.25 mcg/kg bw/day (based on TBTO) or (0.1 mcg/kg bw/day as Sn)

Estimation based on cumulative dietary exposure to TBT##, DBT## and TPT## for average### consumers, while DOT were not taken into account for the estimation of exposure as the EU SCOOP report contains very few data on DOT, which were always below the limit of detection.

international mean concentration of TBT, DBT and TPT of fish and fishery products from European countries participating in the SCOOP task (EFSA 2004) were used for calculation

Country-based "consumer only" total intakes (average intake, (g /day /person, in adults) of fish and seafood products derived from EU reports were used for calculation.

^^ estimated based on intakes of sum of DBT and TBT only from seafood consumption.

Uncertainties and Limitations of the Study

36. While higher accuracy and precision in exposure estimation could be achieved by increasing the sample size, compromise had to be

made due to finite resources. The limited number of samples analysed in the study represents a small fraction of the products available to the local consumers and provided only a snapshot of the levels of OTC in aquatic products.

37. In this study, exposure from other possible OTC sources (e.g. from pesticides, additives used in plastics and other food contact materials) are not included. However, results from studies conducted in other places revealed that fish is an important source of OTC exposure of the general population among different foods and the levels of OTC detected in foods other than seafood were relatively lower.[5]

CONCLUSIONS

38. Among 341 samples of aquatic product, at least one OTC was detected in 205 samples (60%) at quantified levels. The total OTC levels in all samples ranged from below detection level to 490 mcg/kg as Sn. TPT was the most predominant OTC compound. The food group fish was found to be the major contributor of the dietary exposure (88%).

39. The dietary exposures of both average to high consumers of the adult population to the total OTC were below the HBGV. Hence, adverse health outcome due to OTC exposure of the Hong Kong adult population from aquatic products commonly available at local markets was unlikely.

40. The public is advised to maintain a balanced and varied diet which included a wide variety of meat, vegetables and fruits so as to avoid excessive exposure to any contaminants from a small range of food items. As fish contain many essential nutrients, such as omega-3 fatty acids and high quality proteins, moderate consumption of a variety of fish is recommended.

41. Reduction of OTC in aquatic products relies mainly on the control of their use and release in order to safeguard the marine ecosystem and human health. With the increasing number of places to restrict the use of OTC in antifouling paints for ships and pesticides, OTC in the environment are expected to be in a decreasing trend.

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<u>**Appendix I**</u>

Levels of TBT detected in fish, crustaceans and molluscs collected in the current study

Items in each group	No. of samples	Mean TBT levels* LB (mcg/kg)	Mean TBT levels* UB (mcg/kg)	Range of TBT levels* (mcg/kg) LB [UB]
Fish				
Grass carp	8	ND (ND as Sn)	0.61(0.25 as Sn)	—
Salmon	8	ND (ND as Sn)	0.61(0.25 as Sn)	—
Tuna	7	0.64 (0.26 as Sn)	1.2(0.47 as Sn)	ND – 4.5 (ND – 1.8) as Sn [0.61 – 4.5] [0.25 – 1.8] as Sn
Eel	8	ND (ND as Sn)	0.61(0.25 as Sn)	—
Grey mullet	8	ND (ND as Sn)	0.61(0.25 as Sn)	—
Mandarin fish	8	0.23(0.09 as Sn)	0.76(0.31 as Sn)	ND – 1.8 (ND – 0.75) as Sn [0.61 – 1.8] [0.25 – 0.75]as Sn
Chinese noodle fish	8	7.5(3.1 as Sn)	8.0(3.2 as Sn)	ND – 60 (ND – 24)as Sn [0.61 – 60] [0.25 – 24]as Sn
Big head	8	ND (ND as Sn)	0.61(0.25 as Sn)	—
Sea bass	8	0.28(0.11 as Sn)	0.74(0.30 as Sn)	ND – 1.6

				(ND – 0.67)as Sn [0.61 – 1.6] [0.25 – 0.67]as Sn
Seabream	8	0.20(0.08 as Sn)	0.66(0.27 as Sn)	ND – 0.96 (ND – 0.39)as Sn [0.61 - 0.96] [0.25 - 0.39]as Sn
Mackerel	7	0.40(0.16 as Sn)	0.84(0.34 as Sn)	ND – 1.7 (ND – 0.7)as Sn [0.61–1.7] [0.25 – 0.7]as Sn
Pomfret	8	1.04(0.43 as Sn)	1. 3(0.52 as Sn)	ND – 2.6 (0 –1.1)as Sn [0.61 – 2.6] [0.25 – 1.1]as Sn
Cod	8	ND (ND as Sn)	0.61(0.25 as Sn)	–
Sole	8	ND (ND as Sn)	0.61(0.25 as Sn)	–
Turbot	8	ND (ND as Sn)	0.61(0.25 as Sn)	–
Sardine	4	0.70(0.29 as Sn)	1. 2(0.49 as Sn)	ND – 2.8 (ND – 1.2)as Sn [0.61 – 2.8] [0.25 – 1.2]as Sn
Golden thread	8	ND (ND as Sn)	0.61(0.25 as Sn)	–
Horse head	8	ND (ND as Sn)	0.61(0.25 as Sn)	–
Bigeye	8	ND (ND as Sn)	0.61(0.25 as Sn)	–

Mangrove snapper	8	5. 5(2.2 as Sn)	5.6(2.3 as Sn)	ND－24 (ND－10)as Sn [0.61－24] [0.25－10]as Sn
Mud carp	8	ND (ND as Sn)	0.61(0.25 as Sn)	—
Goldfish carp	8	0.10(0.04 as Sn)	0.63(0.26 as Sn)	ND－0.79 (ND－0.32)as Sn [0.61－0.79] [0.25－0.32]as Sn
coral fish	7	0.41(0.17 as Sn)	0.94(0.39 as Sn)	ND－2.9 (ND－1.2)as Sn [0.61－2.9] [0.25－1.2]as Sn
Yellow croaker	8	2.3 (0.95 as Sn)	2.40(0.99 as Sn)	ND－5.1 (ND－2.1)as Sn [0.61－5.1] [0.25－2.1]as Sn
Rabbitfish	8	0.42(0.17 as Sn)	0.80(0.32 as Sn)	ND－1.6 (ND－0.64)as Sn [0.61－1.6] [0.25－0.64]as Sn
Giant grouper	8	0.26(0.10 as Sn)	0.64(0.26 as Sn)	ND－0.8 (ND－0.33)as Sn [0.61－0.8] [0.25－0.33]as Sn
Crustaceans				
Lobster	8	0.43(0.18 as Sn)	0.88(0.37 as Sn)	ND－2.6 (ND－1.1)as Sn [0.61－2.6] [0.25－1.1]as Sn

Mantis shrimp	8	0.54(0.22 as Sn)	0.84(0.35 as Sn)	ND – 2.1 (ND – 0.87)as Sn [0.61 – 2.1] [0.25 – 0.87]as Sn
Shrimp	8	0.50(0.20 as Sn)	1.03(0.43 as Sn)	ND – 4 (ND – 1.7)as Sn [0.61 – 4] [0.25 – 1.7]as Sn
Crab	7	ND (ND as Sn)	0.61(0.25 as Sn)	—
Molluscs				
Scallop	8	0.71(0.29 as Sn)	1.0(0.42 as Sn)	ND – 1.7 (ND – 0.69)as Sn [0.61 – 1.7] [0.25 – 0.69]as Sn
Oyster	8	0.73(0.29 as Sn)	0.96(0.39 as Sn)	ND – 1.9 (ND – 0.79)as Sn [0.61 – 1.9] [0.25 – 0.79]as Sn
Mussel	8	ND (ND as Sn)	0.61(0.25 as Sn)	—
Clam	8	0.95(0.38 as Sn)	1.3(0.52 as Sn)	ND – 2.6 (ND – 1.1)as Sn [0.61 – 2.6] [0.25 - 1.1]as Sn
Geoduck	8	0.45(0.18 as Sn)	0.83(0.34 as Sn)	ND – 1.4 (ND – 0.57)as Sn [0.61 – 1.4] [0.25 – 0.57]as Sn
Razor clam	8	0.60(0.24 as Sn)	0.91(0.37 as Sn)	ND – 1.7 (ND – 0.69)as Sn

				[0.61 − 1.7] [0.25 − 0.69]as Sn
Squid	8	0.09(0.03 as Sn)	0.62(0.25 as Sn)	ND − 0.69 (ND − 0.28)as Sn [0.61 − 0.69] [0.25 − 0.28]as Sn
Octopus	8	1.3(0.52 as Sn)	1.7(0.68 as Sn)	ND − 7.3 (ND − 3)as Sn [0.61 − 7.3] [0.25 − 3]as Sn
Cuttlefish	8	1.2(0.49 as Sn)	1.5(0.62 as Sn)	ND − 3.6 (ND − 1.5)as Sn [0.61 − 3.6] [0.25 − 1.5]as Sn
Sea cucumber	8	ND (ND as Sn)	0.61(0.25 as Sn)	—
Coral clam	8	0.10(0.04 as Sn)	0.63(0.26 as Sn)	ND − 0.76 (ND − 0.31)as Sn [0.61 − 0.76] [0.25 − 0.31]as Sn
Babylon shell	8	0.14(0.06 as Sn)	0.67(0.28 as Sn)	ND − 1.1 (ND − 0.46)as Sn [0.61 − 1.1] [0.25 − 0.46]as Sn
Areolate babylon	8	ND (ND as Sn)	0.61(0.25 as Sn)	—
jade spiral shells	5	ND (ND as Sn)	0.61(0.25 as Sn)	—

***** Rounded to 2 significant figures

Appendix II

Levels of DBT detected in fish, crustaceans and molluscs collected in the current study

Items in each group	No. of samples	Mean DBT levels* LB (mcg/kg)	Mean DBT levels* UB (mcg/kg)	Range of DBT levels* (mcg/kg) LB [UB]
Fish				
Grass carp	8	ND (ND as Sn)	0.49(0.25 as Sn)	—
Salmon	8	ND (ND as Sn)	0.49(0.25 as Sn)	—
Tuna	7	0.10(0.05 as Sn)	0.52(0.26 as Sn)	ND – 0.67 (ND – 0.34)as Sn [0.49 – 0.67] [0.25 – 0.34] as Sn
Eel	8	ND (ND as Sn)	0.49(0.25 as Sn)	—
Grey mullet	8	ND (ND as Sn)	0.49(0.25 as Sn)	—
Mandarin fish	8	ND (ND as Sn)	0.49(0.25 as Sn)	—
Chinese noodle fish	8	0.54(0.28 as Sn)	0.97(0.49 as Sn)	ND – 4.3 (ND – 2.2)as Sn [0.49 – 4.3] [0.25 – 2.2]as Sn
Big head	8	ND (ND as Sn)	0.49(0.25 as Sn)	—
Sea bass	8	0.16(0.08 as Sn)	0.59(0.30 as Sn)	ND – 1.3 (ND – 0.68)as Sn [0.49 – 1.3] [0.25 – 0.68]as Sn

Seabream	8	ND (ND as Sn)	0.49(0.25 as Sn)	—
Mackerel	7	0.12(0.06 as Sn)	0.54(0.27 as Sn)	ND – 0.83 (ND – 0.42)as Sn [0.49 – 0.83] [0.25 – 0.42]as Sn
Pomfret	8	ND (ND as Sn)	0.49(0.25 as Sn)	—
Cod	8	ND (ND as Sn)	0.49(0.25 as Sn)	—
Sole	8	ND (ND as Sn)	0.49(0.25 as Sn)	—
Turbot	8	ND (ND as Sn)	0.49(0.25 as Sn)	—
Sardine	4	0.16(0.08 as Sn)	0.52(0.27 as Sn)	ND – 0.62 (ND – 0.31)as Sn [0.49 – 0.62] [0.25 – 0.31]as Sn
Golden thread	8	ND (ND as Sn)	0.49(0.25 as Sn)	—
Horse head	8	ND (ND as Sn)	0.49(0.25 as Sn)	—
Bigeye	8	ND (ND as Sn)	0.49(0.25 as Sn)	—
Mangrove snapper	8	0.74(0.38 as Sn)	1.05(0.53 as Sn)	ND – 2.9 (ND – 1.5)as Sn [0.49 – 2.9] [0.25 – 1.5]as Sn
Mud carp	8	ND (ND as Sn)	0.49(0.25 as Sn)	—
Goldfish carp	8	ND (ND as Sn)	0.49(0.25 as Sn)	—
coral fish	7	0.09(0.05 as Sn)	0.51(0.26 as Sn)	ND – 0.62

				(ND − 0.32)as Sn [0.49 − 0.62] [0.25 − 0.32]as Sn
Yellow croaker	8	0.08(0.04 as Sn)	0.51(0.26 as Sn)	ND − 0.61 (ND − 0.31)as Sn [0.49 − 0.61] [0.25 − 0.31]as Sn
Rabbitfish	8	ND (ND as Sn)	0.49(0.25 as Sn)	—
Giant grouper	8	ND (ND as Sn)	0.49(0.25 as Sn)	—
Crustaceans				
Lobster	8	0.90(0.46 as Sn)	1.3(0.68 as Sn)	ND − 7.2 (ND − 3.7)as Sn [0.49 − 7.2] [0.25 − 3.7]as Sn
Mantis shrimp	8	ND (ND as Sn)	0.49(0.25 as Sn)	—
Shrimp	8	0.34(0.17 as Sn)	0.71(0.36 as Sn)	ND − 2 (ND −1)as Sn [0.49 − 2] [0.25 − 1]as Sn
Crab	7	ND (ND as Sn)	0.49(0.25 as Sn)	—
Molluscs				
Scallop	8	0.06(0.03 as Sn)	0.49(0.25)	ND − 0.51 (ND − 0.26)as Sn [0.49 − 0.51] [0.25 − 0.26]as Sn
Oyster	8	0.66(0.34 as Sn)	0.78(0.40 as Sn)	ND − 1.4 (ND − 0.71)as Sn [0.49 − 1.4]

				[0.25 − 0.71]as Sn
Mussel	8	1.5(0.76 as Sn)	1.7(0.89 as Sn)	ND − 6.1 (ND − 3.1)as Sn [0.49 − 6.1] [0.25 − 3.1]as Sn
Clam	8	2.3(1.2 as Sn)	2.5(1.3 as Sn)	ND − 6 (ND − 3)as Sn [0.49 − 6] [0.25 − 3]as Sn
Geoduck	8	0.22(0.11 as Sn)	0.59(0.30 as Sn)	ND − 1.1 (ND − 0.56)as Sn [0.49 − 1.1] [0.25 − 0.56]as Sn
Razor clam	8	ND (ND as Sn)	0.49(0.25 as Sn)	—
Squid	8	ND (ND as Sn)	0.49(0.25 as Sn)	—
Octopus	8	0.29(0.15 as Sn)	0.72(0.37 as Sn)	ND − 2.3 (ND − 1.2)as Sn [0.49 − 2.3] [0.25 − 1.2]as Sn
Cuttlefish	8	0.38(0.19 as Sn)	0.69(0.35 as Sn)	ND − 1.5 (ND − 0.76)as Sn [0.49 − 1.5] [0.25 − 0.76]as Sn
Sea cucumber	8	ND (ND as Sn)	0.49(0.25 as Sn)	—
Coral clam	8	2.3(1.2 as Sn)	2. 5(1.3 as Sn)	ND − 8.8 (ND − 4.5)as Sn [0.49 − 8.8] [0.25 − 4.5]as Sn
Babylon shell	8	0.39(0.20 as Sn)	0.64(0.33 as Sn)	ND − 1

				(ND − 0.52)as Sn [0.49 − 1] [0.25 − 0.52]as Sn
Areolate babylon	8	0.06(0.03 as Sn)	0.49(0.25 as Sn)	ND − 0.5 (ND − 0.26)as Sn [0.49 − 0.5] [0.25 − 0.26]as Sn
jade spiral shells	5	ND (ND as Sn)	0.49(0.25 as Sn)	—

* Rounded to 2 significant figures

<u>**Appendix III**</u>

Levels of DOT detected in fish, crustaceans and molluscs collected in the current study

Items in each group	No. of samples	Mean DOT levels* LB (mcg/kg)	Mean DOT levels* UB (mcg/kg)	Range of DOT levels* (mcg/kg) LB [UB]
Fish				
Grass carp	8	ND (ND as Sn)	0.73(0.25 as Sn)	—
Salmon	8	ND (ND as Sn)	0.73(0.25 as Sn)	—
Tuna	7	ND (ND as Sn)	0.73(0.25 as Sn)	—
Eel	8	ND (ND as Sn)	0.73(0.25 as Sn)	—
Grey mullet	8	ND (ND as Sn)	0.73(0.25 as Sn)	—
Mandarin fish	8	ND (ND as Sn)	0.73(0.25 as Sn)	—
Chinese noodle fish	8	ND (ND as Sn)	0.73(0.25 as Sn)	—
Big head	8	ND (ND as Sn)	0.73(0.25 as Sn)	—
Sea bass	8	ND (ND as Sn)	0.73(0.25 as Sn)	—
Seabream	8	ND (ND as Sn)	0.73(0.25 as Sn)	—
Mackerel	7	ND (ND as Sn)	0.73(0.25 as Sn)	—
Pomfret	8	0.11(0.04 as Sn)	0.75(0.26 as Sn)	ND – 0.9 (ND – 0.31)as Sn [0.73 – 0.9]

				[0.25 – 0.31]as Sn
Cod	8	ND (ND as Sn)	0.73(0.25 as Sn)	—
Sole	8	ND (ND as Sn)	0.73(0.25 as Sn)	—
Turbot	8	ND (ND as Sn)	0.73(0.25 as Sn)	—
Sardine	4	ND (ND as Sn)	0.73(0.25 as Sn)	—
Golden thread	8	ND (ND as Sn)	0.73(0.25 as Sn)	—
Horse head	8	ND (ND as Sn)	0.73(0.25 as Sn)	—
Bigeye	8	ND (ND as Sn)	0.73(0.25 as Sn)	—
Mangrove snapper	8	0.38(0.13 as Sn)	0.92(0.32 as Sn)	ND – 1.7 (ND – 0.57)as Sn [0.73 – 1.7] [0.25 – 0.57]as Sn
Mud carp	8	ND (ND as Sn)	0.73(0.25 as Sn)	—
Goldfish carp	8	ND (ND as Sn)	0.73(0.25 as Sn)	—
coral fish	7	ND (ND as Sn)	0.73(0.25 as Sn)	—
Yellow croaker	8	ND (ND as Sn)	0.73(0.25 as Sn)	—
Rabbitfish	8	ND (ND as Sn)	0.73(0.25 as Sn)	—
Giant grouper	8	0.33(0.11 as Sn)	0.96(0.33 as Sn)	ND – 2.6 (ND – 0.89)as Sn [0.73 – 2.6] [0.28 – 0.89]as Sn

Crustaceans				
Lobster	8	ND (ND as Sn)	0.73(0.25 as Sn)	—
Mantis shrimp	8	ND (ND as Sn)	0.73(0.25 as Sn)	—
Shrimp	8	ND (ND as Sn)	0.73(0.25 as Sn)	—
Crab	7	ND (ND as Sn)	0.73(0.25 as Sn)	—
Molluscs				
Scallop	8	ND (ND as Sn)	0.73(0.25 as Sn)	—
Oyster	8	ND (ND as Sn)	0.73(0.25 as Sn)	—
Mussel	8	ND (ND as Sn)	0.73(0.25 as Sn)	—
Clam	8	0.76(0.26 as Sn)	1.1 (0.39 as Sn)	ND – 1.8 (ND – 0.64)as Sn [0.73 – 1.8] [0.25 – 0.64]as Sn
Geoduck	8	ND (ND as Sn)	0.73(0.25 as Sn)	—
Razor clam	8	ND (ND as Sn)	0.73(0.25 as Sn)	—
Squid	8	ND (ND as Sn)	0.73(0.25 as Sn)	—
Octopus	8	ND (ND as Sn)	0.73(0.25 as Sn)	—
Cuttlefish	8	ND (ND as Sn)	0.73(0.25 as Sn)	—
Sea cucumber	8	ND (ND as Sn)	0.73(0.25 as Sn)	—
Coral clam	8	ND (ND as Sn)	0.73(0.25 as Sn)	—

Babylon shell	8	ND (ND as Sn)	0.73(0.25 as Sn)	—
Areolate babylon	8	ND (ND as Sn)	0.73(0.25 as Sn)	—
jade spiral shells	5	ND (ND as Sn)	0.73(0.25 as Sn)	—

***** Rounded to 2 significant figures

Appendix IV

Levels of TPT detected in fish, crustaceans and molluscs collected in the current study

Items in each group	No. of samples	Mean TPT levels* LB (mcg/kg)	Mean TPT levels* UB (mcg/kg)	Range of TPT levels* (mcg/kg) LB [UB]
Fish				
Grass carp	8	0.49(0.17 as Sn)	0.95(0.32 as Sn)	ND – 2.3 (ND – 0.8)as Sn [0.74 – 2.3] [0.25 – 0.8]as Sn
Salmon	8	ND (ND as Sn)	0.74(0.25 as Sn)	—
Tuna	7	130(44 as Sn)	130(45 as Sn)	ND – 850 (ND – 290)as Sn [0.74 – 850] [0.25 – 290] as Sn
Eel	8	0.09(0.03 as Sn)	0.74(0.25 as Sn)	ND – 0.75 (ND – 0.25)as Sn [0.74 – 0.75] [0.25 – 0.25]as Sn
Grey mullet	8	0.31(0.11 as Sn)	0.96(0.33 as Sn)	ND – 2.5 (ND – 0.85)as Sn [0.74 – 2.5] [0.25 – 0.85]as Sn
Mandarin fish	8	0.24(0.08 as Sn)	0.80(0.27 as Sn)	ND – 1.1 (ND – 0.38)as Sn [0.74 – 1.1] [0.25 – 0.38]as Sn

Chinese noodle fish	8	78(26 as Sn)	79 (27 as Sn)	ND－610 (ND－210)as Sn [0.74－610] [0.25－210]as Sn
Big head	8	4.9(1.7 as Sn)	5.2(1.8 as Sn)	ND－29 (ND－9.8)as Sn [0.74－29] [0.25－9.8]as Sn
Sea bass	8	140(47 as Sn)	140 (47 as Sn)	1.1－510 (0.36－170)as Sn [1.1－510] [0.36－170]as Sn
Seabream	8	120(40 as Sn)	120(40 as Sn)	ND－480 (ND－160)as Sn [0.74－480] [0.25－160]as Sn
Mackerel	7	49(16 as Sn)	49(17 as Sn)	ND－200 (ND－67)as Sn [0.74－200] [0.25－67]as Sn
Pomfret	8	45(15 as Sn)	45(16 as Sn)	ND－160 (ND－55)as Sn [0.74－160] [0.25－55]as Sn
Cod	8	3.4(1.1 as Sn)	3.7(1.3 as Sn)	ND－9 (ND－3)as Sn [0.74－9] [0.25－3]as Sn
Sole	8	70(24 as Sn)	70(23.74 as Sn)	1－130 (0.34－44)as Sn [1－130]

				[0.34 – 44]as Sn
Turbot	8	3.8(1.2 as Sn)	4.0(1.3 as Sn)	ND – 11 (ND – 3.7)as Sn [0.74 – 11] [0.25 – 3.7]as Sn
Sardine	4	49(16 as Sn)	49(17 as Sn)	ND – 190 (ND – 65)as Sn [0.74 – 190] [0.25 – 65]as Sn
Golden thread	8	130(45 as Sn)	130(45 as Sn)	91– 170 (31 – 59)as Sn [91 – 170] [31 – 59]as Sn
Horse head	8	77(26 as Sn)	77(26 as Sn)	47 – 120 (16 – 41)as Sn [47 – 120] [16 – 41]as Sn
Bigeye	8	43(15 as Sn)	46(15 as Sn)	ND – 130 (ND – 45)as Sn [0.74 – 130] [0.25 – 45]as Sn
Mangrove snapper	8	450(154 as Sn)	450(154 as Sn)	41 – 1400 (14 – 480)as Sn [41 – 1400] [14 – 480]as Sn
Mud carp	8	0.36(0.12 as Sn)	1.0(0.34 as Sn)	ND – 2.9 (ND – 1)as Sn [0.74 – 2.9] [0.25 – 1]as Sn
Goldfish carp	8	0.23(0.07 as Sn)	0.87(0.30 as Sn)	ND – 1.8 (ND – 0.61)as Sn

				[0.74 – 1.8] [0.25 – 0.61] as Sn
coral fish	7	34(11 as Sn)	34(12 as Sn)	ND – 200 (ND – 68)as Sn [0.74 – 200] [0.25 – 68] as Sn
Yellow croaker	8	90(30 as Sn)	90(30 as Sn)	12 – 200 (4.0 –68)as Sn [12 – 200] [4.2 – 67] as Sn
Rabbitfish	8	33(11 as Sn)	33(11 as Sn)	7.4 – 130 (2.5 – 43)as Sn [7.4 – 130] [2.5 – 43] as Sn
Giant grouper	8	220(75 as Sn)	220(73 as Sn)	11 – 1400 (3.6 – 460)as Sn [11 – 1400] [3.6 – 460] as Sn
Crustaceans				
Lobster	8	1.2(0.41 as Sn)	1.6(0.54 as Sn)	ND – 3.9 (ND – 1.3)as Sn [0.74 – 3.9] [0.25 – 1.3] as Sn
Mantis shrimp	8	140(49as Sn)	140(48 as Sn)	0.92 – 270 (0.31 – 90)as Sn [0.92 – 270] [0.31 – 90] as Sn
Shrimp	8	15(5.2 as Sn)	16(5.6 as Sn)	ND – 110 (ND – 39)as Sn [0.74 – 110] [0.25 – 39] as Sn

Crab	7	ND (ND as Sn)	0.74(0.25 as Sn)	—
Molluscs				
Scallop	8	1.9(0.65 as Sn)	2.1(0.71 as Sn)	ND – 4.5 (ND – 1.5)as Sn [0.74 – 4.5] [0.25 – 1.5] as Sn
Oyster	8	0.11(0.04 as Sn)	0.76(0.26 as Sn)	ND – 0.88 (ND – 0.3)as Sn [0.74 – 0.88] [0.25 – 0.3]as Sn
Mussel	8	ND (ND as Sn)	0.74(0.25 as Sn)	—
Clam	8	280(95 as Sn)	280(95 as Sn)	ND – 700 (ND – 240)as Sn [0.74 – 700] [0.25 – 240] as Sn
Geoduck	8	4.8(1.6 as Sn)	5.1(1.7 as Sn)	ND – 13 (ND – 4.5)as Sn [0.74 – 13] [0.25 – 4.5] as Sn
Razor clam	8	ND (ND as Sn)	0.74(0.25 as Sn)	—
Squid	8	29(9.8 as Sn)	29(10 as Sn)	ND – 140 (ND – 49)as Sn [0.74 – 140] [0.25 – 49] as Sn
Octopus	8	71(24 as Sn)	72(24 as Sn)	ND – 360 (ND – 120)as Sn [0.74 – 360] [0.25 – 120] as Sn
Cuttlefish	8	150(52 as Sn)	150(52 as Sn)	49 – 290

				(17 – 100)as Sn [49 – 290] [17 – 100] as Sn
Sea cucumber	8	ND (ND as Sn)	0.74(0.25 as Sn)	—
Coral clam	8	ND (ND as Sn)	0.74(0.25 as Sn)	—
Babylon shell	8	ND (ND as Sn)	0.74(0.25 as Sn)	—
Areolate babylon	8	47(16 as Sn)	47(16 as Sn)	28 – 72 (9.7 – 25)as Sn [28 – 72] [9.7 – 25] as Sn
jade spiral shells	5	ND (ND as Sn)	0.74(0.25 as Sn)	—

***** Rounded to 2 significant figures

Appendix V

Levels of the sum of the four organotin compounds detected in fish, crustaceans and molluscs collected in the current study

Items in each group	No. of samples	Mean OTC levels*LB (mcg/kg) as Sn	Mean OTC levels*UB (mcg/kg) as Sn	Range of OTC levels (mcg/kg) as Sn LB [UB]
Fish				
Grass carp	8	0.17	1.1	ND – 0.80 [1.0-1.6]
Salmon	8	ND	1.0	–
Tuna	7	45	46	ND – 290 [1.0-290]
Eel	8	0.03	1.0	ND – 0.25 [1.0-1.0]
Grey mullet	8	0.11	1.1	ND – 0.85 [1.0-1.60]
Mandarin fish	8	0.18	1.1	ND – 0.75 [1.0-1.50]
Chinese noodle fish	8	30	31	ND – 240 [1.0-240]
Big head	8	1.6	2.5	ND – 9.8 [1.0-11]
Sea bass	8	48	48	0.36-170 [1.1-170]
Seabream	8	40	41	ND – 160 [1.0-160]
Mackerel	7	17	17	ND – 68

				[1.0-68]
Pomfret	8	16	17	ND－56 [1.0-56]
Cod	8	1.1	2.0	ND－3.0 [1.0-3.7]
Sole	8	24	24	0.34-44 [1.1-45]
Turbot	8	1.3	2.1	ND－3.7 [1.0-4.5]
Sardine	4	17	18	ND－67 [1.0-67]
Golden thread	8	44	45	31-59 [32-60]
Horse head	8	26	27	16-41 [17-42]
Bigeye	8	15	15	ND－45 [1.0-46]
Mangrove snapper	8	160	160	14-490 [15-490]
Mud carp	8	0.13	1.1	ND－1.0 [1.0-1.8]
Goldfish carp	8	0.1	1.0	ND－0.93 [1.0-1.4]
coral fish	7	12	12	ND－68 [1.0-69]
Yellow croaker	8	31	32	4.2-70 [4.9-70]
Rabbitfish	8	11	12	2.5-44 [3.2-44]
Giant grouper	8	73	74	3.6-460 [4.3- 460]

Crustaceans				
Lobster	8	1.1	1.8	ND – 4.8 [1.0-5.3]
Mantis shrimp	8	49	49	0.31-91 [1.1-91]
Shrimp	8	5.8	6.6	ND – 42 [1-42]
Crab	7	0	1.0	—
Molluscs				
Scallop	8	0.97	1.63	ND – 2.2 [1.0-2.7]
Oyster	8	0.67	1.30	ND – 1.3 [1.0-1.8]
Mussel	8	0.77	1.6	ND– 3.1 [1.0-3.9]
Clam	8	97	97	ND – 240 [1.0-240]
Geoduck	8	1.9	2.6	ND – 4.5 [1.0-5.3]
Razor clam	8	0.25	1.1	ND – 0.69 [1.0-1.4]
Squid	8	10	11	ND – 49 [1.0-50]
Octopus	8	24	25	ND – 120 [1.0-120]
Cuttlefish	8	53	53	17-100 [18-100]
Sea cucumber	8	ND	1.0	—
Coral clam	8	1.2	2.0	ND – 4.5

				[1-5.2]
Babylon shell	8	0.26	1.1	ND – 0.98 [1.0-1.5]
Areolate babylon	8	16	17	—
jade spiral shells	5	0	1.0	—

***** Rounded to 2 significant figure