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Organotin contamination in seafood and its implication for human health risk in Hong Kong

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ABSTRACT

Organotins (OTs) have caused widespread adverse effects on marine organisms, while they can also induce health problems to humans via consumption of contaminated seafood. This study aimed to quantify the tissue concentrations of OTs in 11 seafood species in Hong Kong, and assess the human health risk for consuming these species. The tongue sole *Paraplagusia blochii* had the highest concentration of total OTs. Triphenyltin (TPT) accounted for 56–97% of total OTs. The highest hazard quotient (HQ) for TPT was 1.41 in *P. blochii*, while the HQs for butyltins were much less than 1. The results indicated that it is likely to have certain health risks for consuming *P. blochii* due to its high TPT contamination. Therefore, TPT should be a priority pollutant of concern. Appropriate management actions should be taken to control its use and release in the region in order to safeguard the marine ecosystem and human health.

1. Introduction

Organotin compounds (OTs), in particular tributyltin (TBT) and triphenyltin (TPT), are well-known endocrine disruptors and have contaminated our environments for more than 40 years (Yi et al., 2012). Ever since their wide application as biocides in antifouling systems, aquaculture facilities and agriculture starting from the 1960s, OTs have caused widespread adverse effects to many marine and freshwater organisms (Clarke and Smith, 2011). These chemicals have been known to induce the abnormal development of imposex, intersex and female masculinisation in over 260 species of gastropods, and inhibit the growth and development of oysters (Titley-O'Neal et al., 2011). The International Maritime Organization (IMO) has therefore enacted a mandatory global ban on the application of OT-based antifouling systems on all seagoing vessels since September 2008 (IMO, 2008). As a result, it is logical to anticipate that there would be a reduction in OT contaminations in marine waters around the world.

Many marine organisms, however, are still suffering from OT contaminations. As OTs can be easily accumulated in biota as well as along the food chain, marine organisms at higher trophic levels are more susceptible to OTs (Howell and Behrends, 2010). Marine mammals, for example, being the top predator in the marine ecosystem, have higher concentrations of OTs than their prey (Kannan and Falandysz, 1997).

Previous studies showed that OTs are probably able to induce harmful health effects to humans including reproductive and developmental abnormalities, immunosuppression and possible carcinogenic activity (Antizar-Ladislao, 2008). For example, TBT or TPT can inhibit enzyme activity in ovary cells at concentrations as low as 2 ng mL^{-1} (Saitoh et al., 2001) and promote development of prostate cancer cells at 100 nM (Yamabe et al., 2000). Humans are exposed to OTs mainly through three ways namely skin contact, inhalation and ingestion (WHO, 1980). Among them, dietary consumption through contaminated seafood is regarded as the major pathway of OT intake (Yi et al., 2012). These compounds are degradable via bacterial action (i.e., biodegradation) and light irradiation (i.e., photodegradation). However, they cannot be destroyed by cooking (Willemsen et al., 2004). In general, the higher the trophic levels the organisms are, the more OTs are accumulated in their body tissues. Some predatory fishes, such as tuna, salmon, mackerel and cod, are regularly consumed by humans and contribute to nearly 38% of our total OT exposure (Guérin et al., 2007).

Hong Kong has a long history of OT contamination (Ko et al., 1995; Leung et al., 2006; Qiu et al., 2011) although the applications of OT-based antifouling paints on small ships (<25 m in length) and fish cages have been banned since 1992. In the freshwater and marine environments of Hong Kong, these chemicals are still detected in water, sediments and biota. For instance, TBT and its degradation products were detected up to 23.2 μg L^{-1} and 38.6 μg g^{-1} dry weight (dw) in Hong Kong's river waters and sewage sludge respectively (Kueh and Lam, 2008). TBT concentrations in marine sediments could be as high as ca. 130,000 μg kg^{-1} dw

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(53,000 ng Sn g^{-1}; see Ko et al., 1995). In biota, we have recently detected a very high TPT concentration (up to 11,279 μg kg^{-1} dw) in the tissue of the rock shell *Reishia clavigera* (Ho and Leung, unpubl. data). However, we also noted that TBT concentrations in *R. clavigera* were not very high when compared to that of TPT.

Like most coastal cities, Hong Kong has easy accessibility towards fisheries resources and maritime trade. Traditionally, Hong Kong people consume a large amount of seafood in their daily life. According to a food consumption survey performed during 2005–2007, the average consumption rates of fish and molluscs for Hong Kong people were 57.48 g d^{-1} and 5.95 g d^{-1} respectively (CFS, 2010). However, the figures increased to 87.9 g d^{-1} (32.1 kg yr^{-1}) for fish and 52.3 g d^{-1} (19.1 kg yr^{-1}) for molluscs according to the most up-to-date figures documented by Food and Agriculture Organisation (FAO, 2012). These seafood consumption rates ranked among the highest in the world (see Appendix 1). As intake of contaminated seafood is a major pathway of human exposure towards OTs, the human health risk associated with OTs is expected to be high to Hong Kong people.

In a recent report published by the Government of the Hong Kong Special Administrative Region, OTs have been highlighted as one of the top seven endocrine disrupting chemicals of concern in food (CFS, 2012). However, only a few studies reported OT concentrations in seafood (e.g., Harino et al., 2000; Chen et al., 2008). No surveys of OT contamination, including both butyltin (BTs) and phenyltin compounds (PTs), have been conducted in the seafood from Hong Kong and Pearl River Delta region of South China. The associated human health risk concerning OTs from consuming seafood has never been evaluated in this region.

This study, therefore, aimed to (1) investigate, for the first time, the OT tissue concentrations, including both butyltin and phenyltin compounds, in selected seafood species in Hong Kong markets; (2) identify the species or the group of seafood that contains high levels of OTs and (3) perform human health risk assessments to investigate the potential health risk under two estimated seafood consumption rates of Hong Kong people.

2. Materials and methods

This study was designed to measure the tissue concentrations of OTs (i.e., mono-BT, di-BT and TBT, mono-PT, di-PT and TPT) in 11 commonly available seafood species in Hong Kong including three gastropod species (*Babylonia areolata*, *Bufonaria rana* and *Hemifusus tuba*), two bivalve species (*Meretrix lusoria* and *Ruditapes variegatus*) and six fish species (*Collichthys lucidus*, *Harpadon nehereus*, *Johnius belangerii*, *Nibea albiflora*, *Paraplagusia blochii* and *Siganus canaliculatus*). The results were then applied to the human health risk assessment associated with OT-contaminated seafood.

2.1. Sample preparation

2.1.1. Gastropods and bivalves

About 20–40 individuals of each species were bought from two wet markets, namely Tai Po Market (22°26′46.72″, 114°09′59.18″) and Mongkok (22°19′4.4″, 114°10′0.52″), during the wet season of 2012 in Hong Kong. All samples were of marketable size (see Appendix 2). They were iced immediately after purchase and transferred to laboratory. Identification of the animals followed the descriptions and keys in Yang et al. (1992), Shao et al. (1996) and Zhang (2008). For gastropods, we measured the shell length of the gastropods by callipers (to the nearest 0.1 mm). For bivalves, shell width and height were measured instead. Whole body wet weight (with shell) and tissue wet weight were also measured by an electronic balance (to the nearest 1 mg) for all animals. The soft body tissues were taken out for the following analysis.

Imposex identification was conducted for the three gastropod species following the generalised scheme described in Muenpo et al. (2011). Features of their reproductive systems were observed under a stereomicroscope (Olympus SZH10). Penis, if present, was straightened and the length was measured using the scale in the microscopic lens (to the nearest 0.1 mm). We used two imposex indices, namely Vas Deferens Sequence Index (VDSI) and Relative Penis Size Index (RPSI), to access the severity of imposex development. VDSI measures the progressive imposex development by seven stages (0–6). In brief, stage 0 means no imposex developed and stage 6 indicates the most severe stage of imposex. Stages 5 and 6 indicate the infertility of the female due to the blockage of the oviduct opening (Muenpo et al., 2011). RPSI is the fraction between the mean bulk of the female penis and that of the male penis (Gibbs et al., 1987). The bulk of the penis can be expressed as the cube of its length, thus:

$$\text{RPSI} = (\text{Mean length of female penis})^3 \times 100/(\text{Mean length of male penis})^3$$

For each species, five replicates were analysed for tissue concentrations of OTs and each replicate contained a pool of 4–8 individuals.

2.1.2. Fish

Fishes were obtained from two commercial shrimp trawlers operated around western (22°12′14.4″, 113°55′58.8″) and southern Hong Kong waters (22°12′39.6″, 114°17′6″) during the wet season of 2012. Each trawler out-rigger was of 16.9 m. The trawler operated at a speed of 5–7 km h^{-1} for 30 min at each site. Samples were taken from nine replicate nets (beam size: 3.28 m; stretched mesh size: 1.3–2.5 cm). Fish samples were immediately frozen after landing and transferred to laboratory for identification following the descriptions in Shen (1993) and AFCD (2012). Total length, standard length (if applicable) and wet weight were measured for every individual. Only the dorsal muscle was dissected and used for chemical analysis. For large fishes, each individual was treated as a replicate, while for small fishes, several individuals were pooled as a replicate. Five replicates per species were analysed for OTs concentrations. Names of all studied seafood species were checked against and followed the World Registry of Marine Species® (WoRMS®, 2013).

2.2. Chemical analysis

Analysis of BTs and PTs followed the protocol described by Guðmundsdóttir et al. (2011) with slight modifications. Quantification of OTs was performed using a gas chromatograph (GC; Bruker 450-GC, Bruker Inc., Billerica, MA, USA) equipped with a mass-selective detector (Bruker 320-MS, Bruker Inc., Billerica, MA, USA). A VF-5MS fused silica capillary having 0.25 mm i.d. × 30 m × 0.25 m film thickness (Bruker Inc., Billerica, MA, USA) was used as the GC column. The certified reference material ERM-CE477 (mussel tissue) validated the method previously with recoveries of 82–92%. Also, a surrogate standard (di-*n*-heptyltin dichloride) was spiked into each sample to check the recovery. Procedural blanks were analysed simultaneously with each batch of five samples to check for any interference or contamination during the analysis. Limits of detection ranged from 0.2 to 1.5 μg kg^{-1} dw of the six compounds. No correction was made for the recoveries of surrogate standard to the concentrations reported.

All standards were bought from Sigma–Aldrich (St. Louis, MO, USA) and Chiron (Trondheim, Norway). All solvents were in HPLC Grade bought from Tedia (Fairfield, OH, USA).

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