



Accumulation of butyltin compounds in cobia *Rachycentron canadum* raised in offshore aquaculture sites

Shiu-Mei Liu*, Meng-Pei Hsia, Chun-Min Huang

Institute of Marine Biology, National Taiwan Ocean University, 2 Pening Rd., Keelung, Taiwan

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Abstract

Butyltin residues (monobutyltin, MBT; dibutyltin, DBT; tributyltin, TBT; tetrabutyltin, TeBT) in the sea water and in the cobia (*Rachycentron canadum*) from aquaculture sites located offshore of Penhu island, Taiwan, were collected and quantified. The average concentrations of MBT, DBT, TBT and TeBT in sea water were n.d.— 28 ± 3 , 4.0 ± 0.6 – 88 ± 13 , n.d.— 43 ± 4 , and n.d.— 7 ± 1 ng l⁻¹, respectively. The total butyltin (sum of MBT, DBT, TBT, TeBT) residues in the skin, dorsal muscle, ventral muscle, dark muscle, and liver of the cobia were in the range of 72 ± 12 – 2270 ± 85 , 79 ± 11 – 688 ± 33 , 82 ± 14 – 1715 ± 104 , 93 ± 13 – 803 ± 47 , and n.d.— $52,745 \pm 252$ ng g⁻¹ (wet weight), respectively. Although in this study in most cases, the highest concentration of total butyltin residues was found in liver or skin, in some cases, the highest concentration was found in muscle tissue. The crude lipid content in the skin, dorsal muscle, ventral muscle, dark muscle, and liver of these cobia was in the range of 7.9 ± 0.1 – $28 \pm 1\%$, 11.7 ± 0.8 – $29 \pm 1\%$, 11.5 ± 0.3 – $44 \pm 3\%$, 24.2 ± 0.4 – $48.4 \pm 0.4\%$, and 55.7 ± 0.1 – $87.7 \pm 0.4\%$ (wet weight), respectively. The concentrations of crude lipid content, and the concentrations of total butyltin residues in these tissues were not correlated.

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1. Introduction

Organotins are one of the most widely used organometallic compounds. The worldwide production of organotin compounds has increased almost ten times in the past 40 years (Fent, 1996). The major application of organotin compounds (about 70%) is

the use of dialkyltin derivatives as stabilizers in polyvinyl chloride polymers (PVC) processing. Trialkyltin derivatives are used as anti-fouling agent in boat paint and as catalysts in the chemical industry. Due to the wide industrial and agricultural applications considerable amounts of organotins have entered estuarine and marine ecosystems (Hoch, 2001).

Even at very low concentrations (<10 ng l⁻¹), tributyltin (TBT) can cause shell anomalies and failure of spat in oyster (Alzieu et al., 1989), impotence in neogastropods and gastropods (Horiguchi et al., 1997;

* Corresponding author. Tel.: +886 2 2462 2192x5307; fax: +886 2 2463 3152.

E-mail address: smliu@mail.ntou.edu.tw (S.-M. Liu).

Hung et al., 2001) and immunological dysfunction in fish (Suzuki et al., 1992).

Although organotin compounds are suspected of being potent androgen receptor antagonists and that they may cause abnormalities in male reproductive systems (Japan Environment Agency, 1998), little is known about the effects of organotins in humans.

Taiwan has regulated the use of TBT-based paints from the beginning of 2003 and TBT application is prohibited on boats shorter than 25 m in length. However, butyltin compounds can still be encountered in the environment, e.g. TBT leached from anti-fouling paints from big commercial vessels (larger than 25 m in length) and aquaculture nets, and dibutyltin (DBT) leached from PVC pipes/tubes and plastic covers of the floating box used in aquaculture equipments.

Recent studies have shown that organotin levels in the muscle tissue of several fish species intended for human consumption has approached or exceeded the tolerable daily intake (TDI) for humans (Kannan and Falandysz, 1997). Cobia (*Rachycentron canadum*) is one of the main species of fish that is aquacultured in the offshore area of Taiwan. It grows fast and can reach a size of up to about 1.5 m long. Cobia is rich in fats and proteins. Since bioconcentration and bioaccumulation of TBT in living organisms are facilitated by its lipophilic nature, it is necessary to monitor the concentration of butyltin compounds in cobia as a food safety measure.

In this study accumulation and specific distribution of butyltin compounds in various tissues (skin, dorsal muscle, ventral muscle, dark muscle, and liver) of cobia, as well as in sea water collected from the offshore aquaculture sites were determined in order to understand the extent of bioaccumulation or bioconcentration, and to verify if there is a risk for human health.

2. Materials and methods

2.1. Sample collection

Sea water and cobia (*R. canadum*) samples were collected at seasonal intervals from April 2003 to May 2004 from offshore aquaculture sites along Penhu island off the coast of Taiwan (Fig. 1). One sea water

sample and one cobia sample were collected from each of the three aquaculture sites at each sampling time (in some cases less than three cobia were collected). Sea water samples were acidified to pH 4.8 with acetic acid–sodium acetate buffer (1 M) immediately after sampling, and stored in a dark location at 4 °C. The mean body length and body weight of the cobia were determined after sampling. Three subsamples were taken from each water sample (about 100 ml per subsample) and tissue sample, e.g. skin, dorsal muscle, ventral muscle, dark muscle, and liver of the cobia (about 100 g per subsample of each tissue). All the samples were frozen as quickly as possible, and transferred to the laboratory where they were stored at –20 °C.

2.2. Analytical procedures for butyltin compounds

Butyltin compounds in sea water and in different tissues of cobia samples were analyzed as described in previous reports (Hsia and Liu, 2003; Hsia et al., 2004). Fish skin, dorsal muscle, ventral muscle, dark muscle, and liver removed from the cobia were freeze-dried and ground to a homogeneous mixture before analysis. Briefly, the analytical procedure consisted of four steps: (i) acid digestion of the sample; (ii) extraction; (iii) solid phase microextraction (SPME) procedure (Hsia and Liu, 2003) used for sodium tetraethylborate derivatization and extraction to the fiber; (iv) quantitative determination of butyltin compounds by a Dani GC 1000 gas chromatograph equipped with a column (HP-5, 30 m × 0.25 mm i.d. × 0.25 μm film thickness, Hewlett Packard, USA) and a flame photometric detector fitted with a 610 nm optical filter. The analytical procedure for sea water consisted of two steps: (i) solid phase microextraction (SPME) procedure and (ii) quantitative determination of butyltin compounds by a gas chromatograph. For quantification, internal standard tripropyltin chloride was employed and added to the samples prior to extraction. All experiments were conducted in triplicate.

Concentration of butyltin compounds in sea water and in different tissues of cobia were expressed as ng l⁻¹ and ng g⁻¹ on a wet weight basis, respectively. Recoveries of the butyltin compounds from spiked-in-sea water (50 ng l⁻¹, N=6) and in-dorsal muscle of cobia samples (0.3 μg g⁻¹, N=6) ranged from 95–

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