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Total and organic mercury concentrations in the muscles of Pacific albacore (*Thunnus alalunga*) and bigeye tuna (*Thunnus obesus*)

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ABSTRACT

Muscles of 115 North Pacific albacore (ALB, *Thunnus alalunga*) and 75 Pacific bigeye tuna (BET, *Thunnus obesus*), collected from 2001 to 2006, were analyzed. No ALB, but 13 large BET had organic mercury (OHg) concentrations exceeding 1 μg g^{-1} wet weight. For both ALB and BET, total mercury (THg) and OHg concentrations were significantly and positively correlated with fork length (FL) and body weight. The muscle Hg bioaccumulation rates of BET were higher than those of ALB, particularly in the adult fish. Moreover, the lines had crossover points among the two species that imply the young BET (FL < 110 cm) contains lower muscle Hg concentrations than ALB of the same size. The suggested weekly dietary intake of ALB and small-BET meats is 340 g, and of BET meat it is 150 g for a 60-kg person based on the provisional tolerable weekly intake (PTWI) of methylmercury set by the WHO.

1. Introduction

Methylmercury (MeHg), a metal–organic compound, is an environmental toxicant and is produced by microorganisms through biomethylation of mercury (Hg) in the natural environment (Clarkson et al., 2003). High MeHg exposure (Hg poisoning) in humans can cause adverse health effects, for example, the Minamata disease (Harada et al., 1998). Fish is a key food source of Hg exposure in humans (Wheeler, 1996), and MeHg is the major organic compound of Hg in fish (Storelli et al., 2005). Many publications have demonstrated that predator fishes occupying higher trophic levels have higher Hg contaminations because Hg can be biomagnified in predator fishes through marine food chains (Bargagli et al., 1998; Storelli et al., 1998, 2007). Studies have also shown that high Hg exposure in humans is associated with the consumption of contaminated predatory fishes (Storelli et al., 2002, 2005, 2007). Therefore, scientists and health administrators worldwide are concerned about Hg exposure from consuming large predatory fish.

Tunas are migratory predators occupying high trophic positions in marine ecosystems and are important fisheries resources for many nations. Owing to their excellent movement ability, high Hg contamination in tunas is an indicator of non-point global Hg pollution in the ocean. For food safety reasons, Hg contamination in local and imported tuna products has been a concern for many nations (U.S. Food and Drug Administration, 2011; World Health Organization, 2008). Although many publications have reported Hg concentrations in tuna muscle (e.g., Besada et al., 2006; Boush and Thieleke, 1983; Kumar et al., 2004; Menasveta and Siriyong, 1977; Sun and Chang, 1972; Yamashita et al., 2005), relatively fewer studies on total mercury (THg) and organic mercury (OHg) have been reported for North Pacific albacore (ALB) *Thunnus alalunga* and Pacific bigeye tuna (BET) *Thunnus obesus*. In particular, interspecific and intraspecific (gender and size) factors affecting OHg bioaccumulation in the muscles of these two tunas have seldom been studied.

For many marine fishes of high trophic levels, including tunas, sharks and swordfish, studies have shown that muscle Hg contamination relates to their body size (Besada et al., 2006; Menasveta and Siriyong, 1977; Sun and Chang, 1972; Yamashita et al., 2005). For Pacific ALB and BET, positive correlations between muscle Hg concentration and fish size have also been previously reported (Boush and Thieleke, 1983; Morrissey and Geise, 2006;

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Morrissey et al., 2004; Yamashita et al., 2005). However, the sample sizes of previous research have frequently been small, and the ranges of fish size are generally limited to smaller fish. Due to the significant relationship between muscle Hg contamination and fish size, a better understanding of size effects on the muscle Hg contamination of these two tunas would be a benefit to consumers when making decisions about eating tuna meats.

In this study, we analyzed the size effects of Hg bioaccumulation in the muscles of North Pacific ALB and Pacific BET. The aims of this study were to better understand the muscle THg and OHg bioaccumulation patterns of these two commercially important tunas and to provide contamination information about food safety for consumption of these tuna meats. A comparison of the historical data on the muscle Hg concentrations of the two tunas is also presented here.

2. Materials and methods

2.1. Sampling dates, location, and pretreatment

Fish specimens of 115 North Pacific ALB (FL = fork length: 67–118 cm; BW = body weight: 5.9–32.2 kg) and 75 Pacific BET (FL: 81–200 cm; BW: 9–140 kg) were collected by researchers or scientific observers from catches of Taiwanese longline vessels during the periods between October 2001 and April 2006 for ALB and between September 2005 and November 2006 for BET (Fig. 1). For each fish specimen, a white muscle sample was obtained by cutting off a piece of muscle tissue from the dorsal part of the fish, sealing it in a polypropylene bag and then keeping it frozen (<−20 °C) until Hg analysis.

2.2. OHg extraction

For each muscle sample, OHg was extracted and analyzed according to the method used in published papers (Chen and Chou, 2000; Chen et al., 2002, 2007, 2011). Briefly, a subsample (0.2–0.4 g in wet weight) of each muscle sample was weighed in a 40 ml conical graduated centrifuge tube. Acetone was then added to remove lipids covering the surface of the muscle. To extract the lipid phase of Hg, 5 ml of 3 M potassium bromide (KBr) and 10 ml of 0.1 M copper sulfate ($CuSO_4$) were added to the tube. The extractant collected was taken into another conical graduated tube and then extracted again with toluene. The upper organic phase was taken out and further extracted back to 1 ml of 0.01 M sodium persulfate ($Na_2S_3O_3$). This 1 ml $Na_2S_3O_3$ extractant was transferred into a 75 ml graduated test tube for the THg digestion procedure and Hg measurement.

2.3. THg digestion

A subsample (0.2–0.4 g in wet weight) of each white muscle sample was taken into a 75 ml graduated test tube. Subsequently, 1 ml of concentrated nitric acid (HNO_3) and 4 ml of concentrated sulfuric acid (H_2SO_4) were added to the tube. The tube was then heated at 75 °C for 2 h. After cooling down the subsample to room temperature, 11 ml of 5% potassium permanganate ($KMnO_4$) were added for further digestion. Finally, a few ml of 17% H_2O_2 were added to the tube, and the final sample volume of 25 ml was made up with double distilled water.

2.4. Hg determination

Using 5% of tin (II) chloride dehydrate ($SnCl_4$) as a reductant, the muscle Hg concentration of each fish specimen was measured by a cold vapor atomic absorption spectrophotometry (CVAAS, Hitachi Z-8200) with a hydride formation system (Hitachi, HFS-2) attached by a T-joint device (Chen and Chou, 2000). All chemical reagents used in this study were GR grade reagents from Merck Co., Germany. In this study, muscle Hg concentrations were calculated by wet weight. The average wet-to-dry weight ratios (±SD) were 3.40 ± 0.30 for ALB (n = 50) and 3.95 ± 0.40 for BET (n = 60).

2.5. Quality Assurance (QA) and Quality Control (QC)

The certified reference materials used in this study were DORM-2 (dogfish muscle) and DOLT-2 (dogfish liver), purchased from the National Research Council of Canada, and analyzed simultaneously in each batch of the digesting process. For QA and QC, the analytical results of 24 duplicates of each certified reference material (in dry weight) are presented as average ± standard deviation for DORM-2 (THg = 5.57 ± 0.44 and OHg = 3.72 ± 0.28 μg g^{-1}) and for DOLT-2 (THg = 2.58 ± 0.10 and OHg = 0.732 ± 0.05 μg g^{-1}). As compared to the certified values of DORM-2 (THg = 4.64 ± 0.26 and OHg = 4.47 ± 0.32 μg g^{-1}) and DOLT-2 (THg = 2.14 ± 0.10 and OHg = 0.693 ± 0.053 μg g^{-1}), the average values were all within 80% confidence intervals of the certified values.

Moreover, 2 blanks with only digesting reagents were inserted in each digesting process to detect any alien contaminants. For

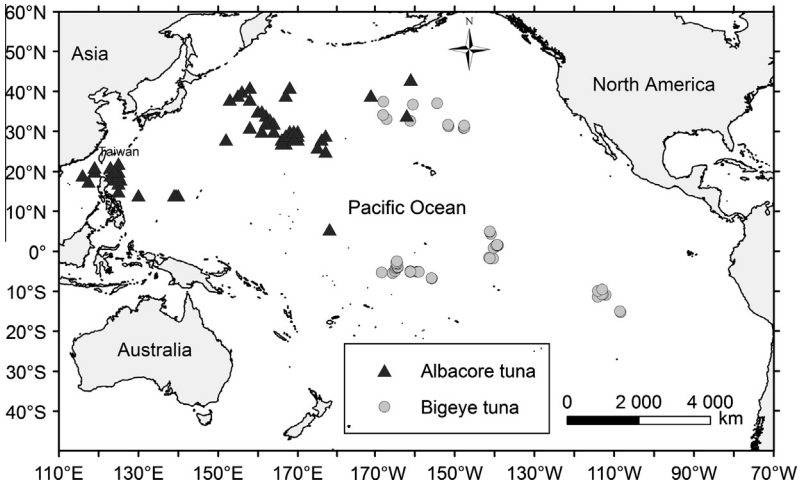

Fig. 1. Map showing the sampling sites for specimens of albacore *Thunnus alalunga* and bigeye tuna *Thunnus obesus* that were collected in the western and central Pacific Ocean from 2001 to 2006.

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