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Tissue-specific distribution of fatty acids, polychlorinated biphenyls and polybrominated diphenyl ethers in fish from Taihu Lake, China, and the benefit-risk assessment of their co-ingestion

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ABSTRACT

The fish tissues from four species collected from Taihu Lake, China, were analyzed including dorsal, ventral, and tail muscles, heart, liver, and kidney. The highest and lowest concentrations of fatty acids were respectively observed in livers and muscles. There were significant intraspecies and interspecies differences in the compositions of most fatty acids among muscle, heart, liver, and kidney. All the tissues were generally beneficial for consumption considering fatty acids. People mainly consume the muscle. Hence, the benefits from two polyunsaturated fatty acids, i.e., eicosapentaenoic acid (EPA) and docosahexaenoic acid (DHA), and risks from PCBs and PBDEs via fish consumption were evaluated by calculating the benefit-risk quotient (BFQ) for the intake of fish muscle containing EPA + DHA vs. PCBs or PBDEs. The BFQ values considering carcinogenic and noncarcinogenic effects for PCBs were ~3000 and 10 times higher than those of PBDEs via fish consumption to achieve the recommended EPA + DHA intake of 250 mg d^{-1}, respectively. The results also suggested that the risk consuming the dorsal muscle was generally lower than the ventral and tail muscles.

1. Introduction

It is known that fish consumption is beneficial to health. The benefits, including reduced risks of coronary heart diseases (CHD), diabetes, and hypertension, are attributed to the presence of long-chain polyunsaturated fatty acids (PUFAs) in fish lipids, especially n-3 fatty acids (which have a double bond starting after the 3rd carbon atom from the methyl end of the carbon chain. Similarly, n-6 fatty acids have a double bond starting after the 6th carbon atom), such as eicosapentaenoic acid (EPA, C20:5n3) and docosahexaenoic acid (DHA, C22:6n3). In contrast, long-chain saturated fatty acids (SFAs) can increase cardiovascular disease risk (Iggman and Riserus, 2011). In addition, certain PUFAs, such as n-6 fatty acids, especially arachidonic acid (20:4n6), exert antagonistic effects on n-3 fatty acids (Luxwolda et al., 2011). Although fish lipids contain all the fatty acids, it is widely believed that fish consumption is beneficial, because fish is generally rich in n-3 fatty acids compared with other types of food.

However, industrialization has led to pollution of the environment and water bodies, and thus contamination of fish. Among the pollutants, persistent organic pollutants (POPs) have been of great concern. POPs are resistant to environmental degradation, are capable of long-range transport, and can be accumulated in organisms and biomagnified through food chains (Wu et al., 2009; Yu et al., 2012). Because of high lipophilicity and persistence, the pollutants can stay in lipids for a long time once they enter into human body, and may cause significant adverse effects on human health. Polychlorinated biphenyls (PCBs), a class of POPs, have been widely used in many industrial applications, especially in capacitors, transformers, and other electrical equipments. Polybrominated diphenyl ethers (PBDEs), another class of POPs, are used in electronics, furniture, and household plastic products as flame retardants. Both of them can be released from the materials and are now ubiquitously present in the environment. PCBs have been listed as the priority-controlled POPs by Stockholm Convention. Penta- and octa-BDE products were nominated as new priority-controlled contaminants listed in Annex A of Stockholm Convention in 2009 (UNEP/POPS/COP.4/17, 2009).

Abbreviations: EPA, eicosapentaenoic acid; DHA, docosahexaenoic acid; PBDE, polybrominated diphenyl ether; PCB, polychlorinated biphenyl.

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Hence, it is important to address whether consumption of contaminated fish is still beneficial to human health. The concentrations of the pollutants as well as the composition profiles and concentrations of fatty acids must be considered. Obviously, the issue depends on the water body where fish lives, the fish species, the amount of fish consumed, the cooking process of fish, and even the different fish tissues. Some authors have made the benefit-risk assessment of co-ingestion of fatty acids and POPs or heavy metals via fish consumption based on the whole muscle of fish (Antonijevic et al., 2007; Sioen et al., 2008; Gladyshev et al., 2009). A few studies have reported the distribution of fatty acids or POPs in fish tissue (Cheung et al., 2008; Chaijan et al., 2010). However, the distribution of both fatty acids and POPs as well as the corresponding estimation of the benefits and risks of co-ingestion of fatty acids and pollutants (both organic toxicants and heavy metals) in the same fish muscles from different portions has not been reported. In China, it is usual that a whole fish with head, bone and skin is sold and consumed. If we know the distributions of both fatty acids and POPs in different types of fish, it may allow consumers to make informed selections about fish consumption. So far, there have been only two papers on the issue about the benefits and risks of co-ingestion of fatty acids and pollutants in China and both considered the whole fish muscles. One reported the benefit-risk assessment of co-ingestion of fatty acids and PCBs in fish from Taihu Lake (Zhang et al., 2012); another reported the benefits and risks of fish consumption from five big cities followed (Du et al., 2012).

Taihu Lake has become the second largest freshwater lake in China because of the shrink of Dongting Lake, the previously largest one. The fish production in Taihu Lake reached 48,000 tons in 2009. In recent years, investigations have been carried out in Taihu Lake, and PCBs and PBDEs were detected in the water body and fish from the lake (Nakata et al., 2005; Liu et al., 2009; Yu et al., 2012). The objective of the present work is to measure the concentrations and compositions of fatty acids, PCBs, and PBDEs in fish collected from Taihu Lake with emphases on (1) the distribution of the compounds in muscles from three portions (i.e., dorsal, ventral, and tail muscle) and viscera (including heart, liver, and kidney that are occasionally consumed by Chinese); (2) the nutritive value assessment of fish tissues via the ratios of PUFA/SFA and n-6/n-3; (3) the muscle-specific benefit-risk estimation via fish consumption considering co-ingestion of fatty acids and pollutants.

2. Material and methods

2.1. Sampling and sample preparation

Fish were caught by commercial fishers from Taihu Lake in September, 2010, as described in our previous study (Zhang et al., 2012). A total of 23 samples covering four species were collected, i.e., silver carp (*Hypophthalmichthys molitrix*), bighead carp (*Hypophthalmichthys nobilis*), common carp (*Cyprinus carpio*), and topmouth culter (*Culter alburnus*), which are the predominant ones in the lake and/or the favorite species for consumption by Chinese. The fish collected were transported to the laboratory in boxes with ice. After measuring the length and weight of each individual fish, they were dissected. Muscles from three portions (i.e., dorsal, ventral, and tail muscle) (Fig. 1), heart, liver, and kidney were obtained. The tissues were homogenized, and parts of the homogenized pastes were used to measure the concentrations and compositions of fatty acids. The remaining pastes of muscles, which would be used to measure the PCB and PBDE concentrations and total lipid contents, were lyophilized to dryness, ground into powders, and stored at −18 °C until the time of use.

2.2. Analytical protocol

Fatty acid concentrations were measured after a two-step treatment, i.e., lipids in the samples were extracted and then trans-esterified as described in our previous study (Zhang et al., 2012). Briefly, wet samples were added to a mixed solvent of chloroform and methanol containing 0.01% butylated hydroxytoluene and the internal standards of nonadecanoic acids and tricosanoic acid (C19:0 and C23:0, Nu-Chek Prep, Inc., USA). The mixtures were subjected to the following steps: vortexed, chloroform added, vortexed, Millipore-water added, and vortexed. Then, the mixtures were centrifugated, and organic phases were dried with anhydrous sodium sulfate and concentrated to nearly dryness. Methanol containing 2% sulfuric acid was added, and the mixtures were heated to 70 °C and held for 2 h. After cooling down, Millipore-water was added, and fatty acid methyl esters (FAMEs) were extracted with *n*-hexane. The extracts were concentrated to 1 mL and stored at 4 °C (<7 days) until analyses.

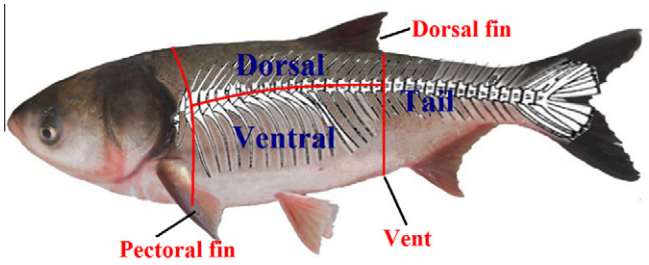

Fig. 1. Muscles from different portions of fish.

The total lipid contents and the concentrations of PCBs and PBDEs were measured according to the protocol described previously (Yu et al., 2011; Zhang et al., 2012). Briefly, the dried samples spiked with the surrogate ^{13}C-PCB141 (Cambridge Isotope Laboratories, USA) were extracted in a Soxhlet extractor by using *n*-hexane:acetone (1:1, v:v) for 72 h. Lipid contents of the samples were determined gravimetrically by using 25% of the extracts. The remaining extracts were cleaned up using concentrated sulfuric acid and a multilayer silica-alumina column as described in our previous studies (Yu et al., 2011; Zhang et al., 2012). Finally, the samples were stored at −18 °C until the time of analysis. In the present study, PCB congeners, including PCB16, 25, 28, 22, 44, 67, 74, 66, 56, 99, 87, 110, 82, 147, 146, 153, 179, 138, 187, 174, 177, 173, 180, 199, 203, 195, 194, and 206, and PBDE congeners, including BDE17, 28, 71, 47, 66, 100, 99, 85, 154, 153, 138, 183, 190, and 209, were quantified.

Analyses of FAMEs were performed on an Agilent 6890N/5975 GC–MS with a DB-WAXETR capillary column (30 m × 0.25 mm × 0.25 μm, J & W Scientific, USA) as described previously (Zhang et al., 2012). Analyses of the PCBs were carried on an Agilent GC/MS operating in electron impact (EI) modes, with A DB-5MS capillary column (60 m × 0.25 mm × 0.25 μm, J & W Scientific, USA). The PBDE congeners were quantified using an Agilent 6890N gas chromatograph (GC) with an HP-5MS capillary column (30 m × 0.25 mm × 0.25 μm, J & W Scientific, USA) coupled to an Agilent 5975 mass spectrometer (MS) under negative chemical ionization (NCI) mode. However, for the analysis of BDE209, an HP-5MS capillary column (12 m × 0.25 mm × 0.25 μm, J & W Scientific, USA) was used. The detailed information was available in our previous studies (Yu et al., 2012; Zhang et al., 2012). The quality assurance and quality control information are provided in the Supporting Information (SI).

2.3. Calculation

The benefit-risk quotient (BRQ) was used to evaluate the benefit-risk of co-ingestion of PUFAs and the pollutants via fish consumption according to the following equation (Gladyshev et al., 2009):

$$BRQ = \frac{CR_{SEFA}}{CR_{lim}}, \quad (1)$$

where CR_{SEFA} (g d^{-1}) is the fish consumption rate to achieve the recommended intake of EPA and DHA (semi-essential fatty acids (SEFA) are fatty acids that humans must ingest because the body requires them for good health and can synthesize them but the synthesis is rather limited. In the present study, SEFA refer to EPA and DHA); CR_{lim} (g d^{-1}) is the maximum allowable fish consumption rate of contaminated fish. If BRQ < 1, to achieve the recommended intake of SEFA, there is no obvious risk to human health via the fish consumption, and vice versa (U.S. EPA, 2000a; Gladyshev et al., 2009).

The CR_{SEFA} is calculated as follows:

$$CR_{SEFA} = \frac{R_{SEFA}}{C_{SEFA}}, \quad (2)$$

where R_{SEFA} (mg d^{-1}) is the recommended SEFA intake rate; C_{SEFA} (mg g^{-1}) is the concentration of SEFA in fish muscle.

The CR_{lim} is calculated as follows (U.S. EPA, 2000a):

$$CR_{lim} = \frac{BW \bullet RfD}{C_p} \quad \text{or} \quad CR_{lim} = \frac{ARL \bullet BW}{C_p \bullet CSF}, \quad (3)$$

where C_p (mg g^{-1}) is the concentration of a pollutant in fish muscle; BW (kg) is the body weight; ARL (unitless) is the maximum acceptable individual lifetime risk level. CSF (mg/kg-d)$^{-1}$ is the cancer slope factor expressed as a cancer potency value of a carcinogen with the unit of risk exposure. RfD (mg (kg-d)$^{-1}$) is an estimate (with