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Fatty acids, polychlorinated dibenzo-*p*-dioxins and dibenzofurans, and dioxin-like polychlorinated biphenyls in paired muscle and skin from fish from the Bohai coast, China: Benefits and risks associated with fish consumption

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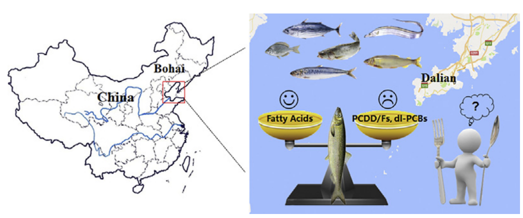
HIGHLIGHTS

- Fatty acid, PCDD/F, and PCB concentrations in fish skin and muscle were determined.
- Total PCDD/F and dl-PCB TEQs were slightly lower for skin than muscle.
- Risks and benefits of consuming fish with and without skin attached were evaluated.

GRAPHICAL ABSTRACT



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ABSTRACT

Fish consumption benefits early cognitive development and cardiovascular health because of the n-3 long-chain polyunsaturated fatty acids in the fish, but toxic pollutants in fish, like dioxin-like polychlorinated biphenyls (dl-PCBs) and polychlorinated dibenzo-*p*-dioxins and dibenzofurans (PCDD/Fs), may decrease or counteract these benefits. In this study, the fatty acids, dl-PCBs, and PCDD/Fs were analyzed in paired muscle and skin from 13 fish and one squid species from the Bohai coast, which have been influenced by serious dioxin pollution from the Bohai Rim Region. The total fatty acid concentrations in the muscle and skin were 2.6–87 and 3.6–156 mg/g wet weight (ww), respectively. The total polyunsaturated fatty acid concentrations were higher in skin than muscle for almost half of the species. The total PCDD/F and dl-PCB toxic equivalents in the muscle and skin were 0.055–0.68 and 0.0099–0.43 pg/g ww, respectively, and were up to five times higher in the muscle than in the skin for eight species. Few benefit–risk studies for fish consumption with and without skin have been performed, so benefit–risk quotients (BRQs) for eating only muscle, only skin, and eating both were calculated. To achieve the recommended 250 mg of EPA + DHA intake, eating only muscle, only skin, and eating both from seven species with BRQs < 1 would cause no significant risk. Removing skin before eating is not necessary for these species. Three fish species had BRQs < 1 for muscle only but BRQs > 1 for skin only. Removing skin before eating would be better for these species. The other four fish species had BRQs > 1 for eating only muscle, only skin, and eating both, indicating the potential risk caused by PCDD/F and dl-PCB. Amounts of the four fish species consumed should be decreased, and other sources of EPA and DHA should be selected.

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1. Introduction

Eating fish offers health benefits. Fish contains high-quality protein, iodine, selenium, and phosphorus, and, above all, is an important source of fatty acids such as n-3 long-chain (n-3 LC) polyunsaturated fatty acids (PUFAs) (Yamada et al., 2014). The n-3 LC-PUFAs are unsaturated fatty acids each with a double bond at the third carbon atom from the methyl end. Some n-3 LC-PUFAs, such as 18:3 alpha-linolenic acid (ALA), 20:5 eicosapentaenoic acid (EPA) and 22:6 docosahexaenoic acid (DHA), are essential fatty acids that humans cannot synthesize or cannot synthesize in the amounts required (Mahaffey et al., 2011). It has been well documented that essential fatty acids, particularly EPA and DHA, protect against cardiovascular disease, autoimmune and neurodegenerative diseases, infant and nervous system development disorders, and even some cancers (Cohen et al., 2005; Hibbeln et al., 2007; Paliwoda et al., 2016; Riediger et al., 2009). Fish, particularly oily fish, are richer than other foods in n-3 LC-PUFAs, so health organizations recommend that a certain amount of fish be consumed every week (American Heart Association, 2010; Chinese Nutrition Society, 2016; Health Canada, 2002; USFDA, 2004). However, due to the widespread water contamination, fish has been found to accumulate xenobiotic compounds, such as persistent organic pollutants (POPs), which cause diseases and can counteract the health benefits of n-3 LC-PUFA in fish. Polychlorinated dibenzo-*p*-dioxins and polychlorinated dibenzofurans (PCDDs and PCDFs, respectively, or PCDD/Fs together) and dioxin-like polychlorinated biphenyls (dl-PCBs) are one kind of the most notorious POPs. They have been classified as probable human carcinogens by the International Agency for Research on Cancer (IARC), and can also cause immunosuppression and behavioral changes and increase the risk of cardiovascular disease in humans and animals (Birnbaum and Tuomisto, 2000; Hojo et al., 2002; IARC, 1997; IARC, 2012; Lind et al., 2004; Vreugdenhil et al., 2002). PCDD/Fs and dl-PCBs are lipophilic, so tend to be deposited in fatty tissues in fish. PCDD/F and dl-PCB concentrations are likely to be higher in fish rich in n-3 LC-PUFA (e.g., oily fish, which health organizations recommend for consumption) than in fish with lower n-3 LC-PUFA contents.

The presence of fatty acids and PCDD/Fs and dl-PCBs in fish brings much confusion for fish consumers. Risk–benefit assessments for the co-ingestion of fatty acids and PCDD/Fs and dl-PCBs have been performed in several studies to allow scientifically based practical advice on fish consumption to be developed (Dewailly et al., 2007; Domingo et al., 2007; Gao et al., 2015; Lundebye et al., 2017; Szlinder-Richert et al., 2011; Usydus et al., 2009). However, the conclusions affirmed by these studies, whether the benefits of fish consumption were greater than the potential risks, were inconsistent. It is generally accepted that the geographical source of fish affects contaminant concentrations and the risk–benefit balance for consumption (Lundebye et al., 2017; Strandberg et al., 2016). Fish from close to industrial areas are likely to have higher contaminant concentrations than other places. Eating fish from near areas with intensive anthropogenic activities is therefore of particular concern, and the risk–benefit balance of eating such fish should be assessed. The Bohai Sea in north China is surrounded by the Bohai Rim Region, one of the three most densely industrialized zones in China. Historical activities of chlorine based chemical industries in this region, like the pentachlorophenol factories, caused severe PCDD/F and dl-PCB pollution of the surroundings, which further posed a long-term impact on the Bohai Sea (Hu et al., 2005; Li et al., 2013; Li et al., 2012). Due to their lipophilicity and resistance to degradation, PCDD/Fs and dl-PCBs from the historical pollution could enter into food chain and accumulate in the fish. The Bohai Sea is also known as the 'fish storehouse' of China, and provides fish to cities nearby and across China (Zhang et al., 2009). Previous studies investigating the PCDD/F and dl-PCB concentrations in seafood from the Bohai Sea have mainly focused on the risks posed by contaminants, but the benefits of consuming seafood have not been fully considered (Shen et al., 2009; Wan et al., 2005; Yang et al., 2009; Zhao et al., 2005). When considering public health, the risks of consuming fish from the Bohai Sea need to be studied together with the benefits, so a full risk–benefit assessment should be performed.

Additionally, most previous studies of the risks and benefits of fish consumption have been for fish fillets with or without the skin attached; so far, the effects of removing the skin have not been assessed. It has been suggested in some risk assessment studies that removing the skin would decrease contaminant intakes, but the decrease will be different for different fish of the same species, for different species, and for different contaminants (Dellinger et al., 1995; Foran et al., 2005a; Hora, 1981; Zabik et al., 1996; Zhang et al., 2013). From the health benefit perspective, fish skin has been shown to be nutritionally valuable, perhaps even more valuable than the flesh in terms of healthy lipids (Sahena et al., 2010; Wang et al., 2016; Zuta et al., 2003). The nutritional benefits of and possible risks posed by eating fish fillets with and without the skin attached should be assessed to determine whether the skin should be removed before fish is consumed. In several traditional cultures, especially Chinese cuisine, fish skin is a delicacy and specialty that is often eaten fried, in salad, or jellied. Benefit-risk assessments for fish skin consumption therefore need to be performed for certain groups.

In this study, we determined fatty acid (particularly n-3 LC-PUFA), PCDD/F, and dl-PCB concentrations in paired muscle and skin samples from marine fish from the coast of the Bohai Rim Region. The objectives were (1) to determine the concentrations of fatty acids and pollutants in fish muscle and skin and assess the risks and benefits of consuming fish from the Bohai Sea, and (2) to determine whether fish should be consumed with the skin removed or attached from the perspective of the risks and benefits. The results will provide reference data to allow public health officials and fish consumers to make appropriate choices regarding fish consumption to minimize the risks posed by the presence of contaminants while maximizing the benefits from nutrients in fish.

2. Materials and methods

2.1. Sample collection

A total of 93 fish (from 13 species) and three squid samples (of one species) were collected from the Dalian coast in May 2014. The species were Pacific cod (*Gadus microcephalus*), bastard halibut (*Paralichthys olivaceus*), ditrema (*Ditrema temminckii*), shishamo (*Spirinchus lanceolatus*), largehead hairtail (*Trichiurus lepturus*), small yellow croaker (*Larimichthys polyactis*), large yellow croaker (*Larimichthys crocea*), Pacific saury (*Cololabis saira*), Japanese halfbeak (*Hyporhamphus sajori*), South American pilchard (*Sardinops sagax*), Pacific herring (*Clupea pallasii*), Japanese Spanish mackerel (*Scomberomorus niphonius*), Japanese seabass (*Lateolabrax japonicas*), and squid (*Loligo japonica*). Biometric information and biological characteristics of the samples are given in Table S1. All the species analyzed are commonly consumed by people living near the Bohai Sea. Once collected, the samples were placed in an ice chest and transported to the laboratory. The head, scales, tail, fins, and guts were removed from each fish and the belly flap was trimmed off. The fish was then boned to yield fillets. The internal organs, arms, and tentacles of each squid were discarded and the mantle was used to obtain fillets. For each fillet, the skin and subcutaneous fat (referred to collectively as skin) were carefully separated from the muscle. The skin and muscle tissue samples from each fillet were then pooled with skin and muscle samples from other individuals to form composite samples for each species. The composite samples were freeze-dried, pulverized, homogenized, and stored in amber glass bottles at −20 °C until required for extraction.