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# Evaluation of human health risks posed by carcinogenic and non-carcinogenic multiple contaminants associated with consumption of fish from Taihu Lake, China

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ABSTRACT

The present study estimated the human daily intake and uptake of organochlorine pesticides (OCPs), polychlorinated biphenyls (PCBs), polybrominated diphenyl ethers (PBDEs), polycyclic aromatic hydrocarbons (PAHs), and toxic trace elements [mercury (Hg), chromium (Cr), cadmium (Cd), and arsenic (As)] due to consumption of fish from Taihu Lake, China, and the associated potential health risks posed by these contaminants. The health risks posed by the contaminants were assessed using a risk quotient of the fish consumption rate to the maximum allowable fish consumption rate considering the contaminants for carcinogenic and non-carcinogenic effect endpoints. The results showed that fish consumption would not pose non-cancer risks. However, some species would cause a cancer risk. Relative risks of the contaminants were calculated to investigate the contaminant which posed the highest risk to humans. As a result, in view of the contaminants for carcinogenic effects, As was the contaminant which posed the highest risk to humans. However, when non-carcinogenic effects of the contaminants were considered, Hg posed the highest risk. The risk caused by PBDEs was negligible. The results demonstrated that traditional contaminants, such as As, Hg, DDTs (dichlorodiphenyltrichloroethane and its metabolites), and PCBs, require more attention in Taihu Lake than the other target contaminants.



## 1. Introduction

With the rapid development of industry and agriculture in China over the past 30 years, many organic and inorganic pollutants have been released into the environment. The release of contaminants into the environment poses a serious threat to biological and human life. Of the organic pollutants, there are a series of chemicals which have persistent potential and are capable of long-range transport via air, water, and species migration. One particular type of the most notorious organic contaminants are the persistent organic pollutants (POPs), such as organochlorine pesticides (OCPs), polychlorinated biphenyls (PCBs), polybrominated diphenyl ethers (PBDEs), and polycyclic aromatic hydrocarbons (PAHs). Due to the high lipophilicity and low degradation capacity of these chemicals, POPs can generally accumulate in organisms and are biomagnified through food chains with elevated levels in top predators (Takeuchi et al., 2009; Wang et al., 2012a; Yu et al., 2012b). These chemicals have significant adverse effects on organisms. Some of these contaminants were therefore listed as priority-controlled pollutants by the Stockholm Convention and the United States Environmental Protection Agency (USEPA). With regard to inorganic pollutants, some substances, such as toxic trace elements, are also persistent in the environment, and some such as mercury (Hg), are biomagnified through food chains (Nfon et al., 2009; Tadiso et al., 2011). Exposure to toxic trace elements, even at low concentrations, over a long period of time is harmful to organisms (Thomas et al., 2009; Zheng et al., 2011).

Taihu Lake, which has an area of 2250 km$^2$ and an average depth of 2 m, is the second largest freshwater lake in China. In the past three decades, industrial, agricultural, and economic development around Taihu Lake has seen tremendous growth. Many contaminants have been detected in the water system of the lake (Liu et al., 2009; Qiu et al., 2010; Wang et al., 2012a; Yu

*Abbreviations:* ARL, the maximum acceptable individual lifetime risk level; B[a]P, benzo[a]pyrene; CSF, cancer slope factor; DDT, dichlorodiphenyltrichloroethane; EDI, estimated daily intake; EDU, estimated daily uptake; HCH, hexachlorocyclohexane; HC, halogenated compound; RQ, risk quotient; OCP, organochlorine pesticide; PAH, polycyclic aromatic hydrocarbon; PBDE, polybrominated diphenyl ether; PCB, polychlorinated biphenyl; RfD, reference dose; RR, relative risk; TEF, toxic equivalency factor.

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et al., 2012b). One of the primary concerns of water systems, especially those in close proximity to urban or/and industrial environments, is the potential health effects of contaminants to humans via the consumption of fish, shellfish, and other biological species from the water bodies. Because of the widespread existence of contaminants, potential human health risks via contaminated fish consumption exist universally. Fish are reliable indicators of the bioaccumulation of persistent toxic substances in the environment, and have been used to estimate contaminant exposure risks to humans (Meng et al., 2007; Yu et al., 2011, 2012b). There is a thriving fishery in Taihu Lake, and the estimated fish production reached 45,000 tons in 2013. If the fish consumption rate was set at 227 g/meal for an average city resident (USEPA, 2000), it would affect the health of approximately two hundred million populations, considering the residents consumed the fish only once. The assessment of human risk via fish consumption from the lake is therefore an important issue, while there was very limited information available.

Contaminants can be classified as carcinogen and non-carcinogen, and they generally coexist in fish. For carcinogens, they can cause both carcinogenic and non-carcinogenic effects on organisms. Therefore, there are two data of the maximum allowable fish consumption rates considering the two effect endpoints. As suggested by USEPA, to protect human health, the lower one of the maximum allowable fish consumption rate considering either carcinogenic or non-carcinogenic effect should be used. The USEPA has therefore developed human health risk assessment methods for carcinogenic and non-carcinogenic contaminants associated with fish consumption (USEPA, 2000). Despite the growing public interest in multiple contaminants, there are limited studies assessing human health risks via fish consumption considering multiple contaminants for both carcinogenic and non-carcinogenic effects (Wang et al., 2013). One of the reasons is that most contaminants have multiple end point effects and it has been challenging to conduct a proper risk assessment for a chemical mixture. Another important reason might be the equations to calculate the maximum allowable fish consumption rate ($CR_{lim}$, an allowable daily consumption of contaminated fish based on a contaminant's carcinogenicity or non-carcinogenic effects) considering multiple contaminants for non-carcinogenic effects is incorrect in the USEPA guideline, because the more pollutants in fish are, the more fish we can eat calculated on the basis of the equation (USEPA, 2000; Yu et al., 2012a). After the correction of the equation, the assessment considering multiple contaminants for non-carcinogenic effects is possible (Yu et al., 2012a; Wang et al., 2013). The main objective of the present study was therefore to assess the potential human health risks posed by the consumption of fish collected from Taihu Lake in September, 2009, considering multiple contaminants for both carcinogenic and non-carcinogenic effects.

## 2. Methodology of risk assessment

### 2.1. Target substances and hazard identification

The toxic substances subject to the risk assessment were those measured in the monitoring campaign in September 2009, as part of the program of the Water Environmental Quality Evolution and Water Quality Criteria in Lakes (the National Basic Research Program of China). Fish was caught by commercial fishers from Taihu Lake. During the field campaign, we collected a total of 200 samples (over 1346 individuals) of 24 fish species, which are the generally collected species and they accounted at least over 90% of the productions. We measured the concentrations of OCPs, PCBs (39 PCB congeners, i.e., PCB1, 2, 3, 4, 6, 8, 9, 16, 18, 19, 22, 25, 28, 44, 52, 56, 66, 67, 71, 74, 82, 87, 99, 110, 138, 146, 147, 153, 173, 174, 177, 179, 180, 187, 194,195, 199, 203, and 206), PBDEs (14 PBDE congeners, i.e., BDE17, 28, 47, 66, 71, 85, 99, 100, 138, 153, 154, 183, 190, and 209), PAHs (15 PAHs, i.e., acenaphthylene, acenaphthene, fluorene, phenanthrene, anthracene, fluoranthene, pyrene, benz[a]anthracene, chrysene, benzo[a]pyrene, benzo[b]fluoranthene, benzo[k]fluoranthene, indeno[1,2,3-c,d]pyrene, dibenz[a,h]anthracene, and benzo[g,h,i]perylene), synthetic musks, and trace elements [including iron (Fe), manganese (Mn), zinc (Zn), mercury (Hg), chromium (Cr), cadmium (Cd), lead (Pb), and arsenic (As)] in the fish species collected, and investigated the bioaccumulation and biomagnification behavior of these substances through the food chain in Taihu Lake (Hao et al., 2013; Wang et al., 2012a; Yu et al., 2012b; Zhang et al., 2013). In the present study, for OCPs, only DDTs and HCHs (hexachlorocyclohexanes) were considered as these two types of chemicals contributed 90.2% (mean) of the total OCPs (Wang et al., 2012a). The trace elements, Fe, Mn, and Zn, were not considered because these elements are recognized as human essential elements, although excess exposure in organisms to such elements may result in toxic effects (Gopalani et al., 2007). No RfD (reference dose) and CSF (cancer slope factor) values for synthetic musks and Pb were available in the Integrated Risk Information System (IRIS) of the USEPA (http://www.epa.gov/IRIS/), therefore, these substances were not considered. The other substances were assessed in terms of risks to humans following consumption of the fish species collected. In addition, because only 1–2 samples of some of the 24 fish species were collected, the species in which the numbers of samples was more than 5 were used in the present study. As a result, only 16 types of fish species were used. They were one types of herbivorous fish, i.e., Wuchang bream (*Megalobrama amblycephala*); eight types of omnivorous fish, i.e., crucian carp (*Carassius carassius*), common carp (*Cyprinus carpio*), silver carp (*Hypophthalmichthys molitrix*), Sharpbelly (*Hemiculter leucisculus*), largefin bitterling (*Acheilognathus macropterus*), bighead carp (*Hypophthalmichthys nobilis*), garfish (*Hyporhamphus intermedius*), and yellow catfish (*Pelteobagrus fulvidraco*); and seven types of carnivorous fish, i.e., tapertail anchovy (*Coilia ectenes taihuensis*), spotted steed (*Hemibarbus maculates*), redfin culter (*Cultrichthys erythropterus*), topmouth culter (*Culter alburnus*), clearhead icefish (*Protosalanx hyalocranius*), icefish (*Neosalanx taihuensis Chen*), mongolian redfin (*Chanodichthys mongolicus*).

The hazard identifications of the toxic substances were mainly based on the IRIS (http://www.epa.gov/IRIS/). Among the contaminants, As (inorganic) is classified as group A – known human carcinogen; PCBs, α-HCH, DDTs, and Cd are classified as group B – probable human carcinogen; Hg (methylmercury, MeHg) and β-HCH are classified as group C – possible human carcinogen; pentabromodiphenyl and octabromodiphenyl ethers (penta- and octBDEs), δ-HCH, Hg (elemental), and Cr (VI) are classified as group D – not classifiable as to human carcinogenicity; and no information about the classification for decaBDE and γ-HCH in humans was available on the carcinogenicity. However, for PAHs, some congeners belong to group B, such as benzo[a]pyrene (B[a]P), benz[a]anthracene, benzo[b]fluoranthene, and benzo[k]fluoranthene, and some are classified as group D, such as acenaphthylene and benzo[g,h,i]perylene, and to date, there is no available data for some others, such as acenaphthene.

### 2.2. Dose–response assessment

A dose–response assessment is one of the most important sections of human health risk assessment. The risk endpoints of contaminants in humans are divided into carcinogenic and non-carcinogenic effects. Carcinogenic risk is estimated as the incremental probability of an individual developing cancer over a lifetime as a result of exposure to a potential carcinogen, and assumed to be proportional to cumulative exposure and, at low exposure levels, may be very small or even zero. The CSF, derived from dose–response data obtained in epidemiological studies or chronic animal bioassays, is used to assess an upper-bound probability of an individual developing cancer as a result of a lifetime (usually 70 years) exposure to a particular level of the potential carcinogen (USEPA, 2000). The non-carcinogenic effect of a toxic substance is considered to be acute exposure effects over brief periods of time, such as hours or days. However, this does not necessarily result in an acute response. The RfD is an estimate of daily exposure in humans that is likely to be without an appreciable risk of deleterious effects during a lifetime. The CSF and RfD values of the target contaminants from USEPA (2000) and IRIS (http://www.epa.gov/IRIS/) are listed in Table 1.

### 2.3. Exposure assessment

Human exposure assessment of a contaminant through oral ingestion is generally estimated using daily intake and uptake of the contaminant. Uptake of a substance is defined as the intake multiplied by the uptake efficiency of the substance in the human gastrointestinal tract. In the present study, the uptake efficiencies of the contaminants, which were estimated using the contaminant bioaccessibility measured via in vitro test simulating the human gastrointestinal tract, were cited from literature or calculated based on the literature equations (Table 1). The bioaccessible fraction is considered to be the maximal amount available for absorption in the human gastrointestinal tract. The estimated total daily intake (EDI) and uptake (EDU) of the contaminants in a given fish species were calculated as follows:

$$\mathrm{EDI} = \frac{R_{\mathrm{fish}}}{\mathrm{BW}} \sum_{m=1}^{x} C_m (m = 1, 2, \ldots, \text{and } x) \quad (1)$$

$$\mathrm{EDU} = \frac{R_{\mathrm{fish}}}{\mathrm{BW}} \sum_{m=1}^{x} (C_m \cdot \mathrm{AR}_m)(m = 1, 2, \ldots, \text{and } x) \quad (2)$$