Persistent Organic Pollutants in Aquaculture Systems in the Pearl River Delta, with Focus on Their Bioaccessibility via Fish Consumption

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This study aimed to characterize the distribution of PAHs and OCPs in fish muscle and sediments collected from freshwater fishponds and mariculture zones in the Pearl River Delta (PRD); evaluate the enrichment of PAHs and OCPs in mariculture sediments; study the dietary intake and human health risks of PBDEs and their structural analogues (MeO-BDEs, OH-BDEs and BRPs) via fish consumption; and investigate the bioaccessibilities of PAHs and OCPs via fish consumption.

The concentrations of PAH, HCHs and DDTs in surface sediments collected from the freshwater fishponds and mariculture zones in the PRD ranged from 52.7 to 717 ng g⁻¹, from N.D. to 44.4 ng g⁻¹, and from N.D. to 20.4 ng g⁻¹, dw, respectively. Generally, the PAH and OCP contamination levels in the aquaculture sediments of the PRD were moderately high when compared with other parts of China and worldwide. Our results revealed that the source of PAHs was mainly derived from mixed combustion (66.2%) in freshwater fishpond sediments. As for mariculture sediments, vehicle emissions and coal combustion were the main sources (52.5% and 47.5% of ∑PAH, respectively) of PAHs.

In fish muscle collected from the aquaculture systems, the concentrations of PAHs, HCHs and DDTs ranged from 10.2 to 24.1 ng g⁻¹, from 0.15 to 4.38 ng g⁻¹, and from 0.78 to 12.3 ng g⁻¹, ww, respectively. The percentages exceeding the screening values (SV) of PAHs, HCHs and DDTs were 61.2%, 1.65% and 4.1% in freshwater fish, and 83.6%, 64.9% and 17.6% in marine fish, respectively. A more conservative maximum consumption rate (69 g day⁻¹, the average of CRmax of all fish species) was recommended in order to reduce the cancer risks caused by PAHs, HCHs and DDTs via fish consumption.

In comparison with the sediments from the corresponding reference sites, the average enrichment percentages for TOC, PAHs, HCHs and DDTs were 21.4%, 43.8%, 34.7% and 676% in surface aquaculture sediments, and 24.6%, 73.7%, 21.9% and 1296% in core aquaculture sediments, respectively. The enrichment percentage could be even as high as 8663% for o, p' ‐DDD. Significant positive correlations between the proportions of individual PAHs, HCHs and DDTs (excluding p, p ‐DDD) in trash fish with that in surface mariculture sediments
suggested that fish feeds were the main sources of the enriched POPs in mariculture sediments. Another important source for the enriched DDTs, especially $p, p'$-DDD, was in the form of dechlorinated DDTs released from antifouling paints under anaerobic conditions. Results of ecological risk assessments revealed that enriched OCPs had high potential to contaminate surrounding marine environment leading to adverse effects on the associated biota.

Twenty-two PBDEs, 7 MeO-BDEs, 15 OH-BDEs and 3 bromophenols (BRPs) were analyzed in twenty fish species (279 samples). The estimated daily intakes of PBDEs, MeO-BDEs, OH-BDEs and BRPs via fish consumption ranged from 4.4 to 14, 0.50 to 4.3, 0.02 to 0.43 and 0 to 0.21 ng/kg-day for Hong Kong residents, respectively, based on 50th and 95th centile concentrations. BDE-47 and 99 were found to be the major PBDE congeners while 2'-MeO-BDE-68, 6-MeO-BDE-47 and 3-MeO-BDE-47 were the dominant MeO-BDEs. Concentrations of OH-BDE and BRP were 10 to 100-fold less than those of PBDEs, with small frequencies of detection (max 36.7%). Dietary intake of PBDEs via fish consumption by Hong Kong residents was greater than many developed countries, such as the USA, UK, Japan and Spain. The results indicated that the potential toxicity of these compounds should not be neglected.

An in vitro gastrointestinal digestion model was performed to evaluate the bioaccessibility of PAHs, HCHs and DDTs in twenty fish species collected from Hong Kong markets. Based on the model, the average bioaccessibilities were 24.3, 31.1% in gastric and intestinal conditions for ΣPAHs, 3.35, 8.73% for ΣHCHs and 5.48, and 17.6% for ΣDDTs, respectively. Significant (p<0.05) correlations were observed between OCP congener digestible concentrations in fish muscle and their corresponding concentrations in human tissues of Hong Kong residents. The results suggested that food (such as fish) intake was the predominant pathway for the body loadings of PAHs and OCPs. It was concluded that human health risk assessment based on solvent concentrations should be modified by taking bioaccessibility of the contaminant into account because only a limited proportion of OCP was bioaccessible.

The present study revealed that the fish and sediments collected from the freshwater fishponds and mariculture zones of the PRD were moderately contaminated by PAHs and OCPs. Reducing POPs contamination of fish feeds is recommended because fish feeds contribute significantly to the contamination of PAHs and OCPs in fish muscle as well as their enrichment in
mariculture sediments. Furthermore, new emerging chemicals such as MeO-PBDEs and OH-PBDEs in fish samples should not be neglected for their potential toxicity. The results also suggested that risk assessment based on solvent concentrations might over estimate the human health risks.
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