Persistent halogenated compounds in aquaculture environments of South China: Implications for global consumers’ health risk via fish consumption

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ARTICLE INFO

Article History:
Received 11 January 2011
Accepted 20 April 2011
Available online 13 May 2011

Keywords:
Persistent halogenated compounds
Input sources
Uptake routes
Health risk
Monte Carlo simulation

ABSTRACT

This study examined the potential sources of persistent halogenated compounds (PHCs), including organochlorine pesticides, mainly DDXs (sum of o,p′- and p,p′-DDT, -DDD, and -DDE and p,p′-DDMU) and polybrominated diphenyl ethers and to typical aquaculture environments of South China, determined the relative importance of gill diffusion and fish feeding for exposure of fish to these contaminants and assessed potential health risk for global consumers via consumption of fish from South China. Fish feed is generally a direct and important source of PHCs in both freshwater and seawater aquaculture. In addition, gill diffusion is the predominant uptake route for PHCs (except p,p′-DDMU, o,p′-DDD and -DDE) in farmed freshwater fish, whereas accumulation from the diet is the major route for farmed marine fish. Risks to health of global consumers via consumption of fish from South China are minimal. However, increased risk can be foreseen due to continuous use of brominated fire retardants and electronic waste importation to China.

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1. Introduction

Global aquaculture has developed rapidly in recent years, growing at an annual rate of 6.4% from 2002 to 2006, and has been the fastest growing sector among all the animal-based food industries. It was reported that fish provide at least 15% of animal protein intake per capita for more than 2.9 billion people (Food, Agriculture Organization of the United Nations, 2009). In addition, aquaculture in developing countries will continuously grow in the next 10 years, with seafood consumption and production accounting for 77% and 79%, respectively of the global totals (Food, Business Network, 2008). China, as the world’s largest fishery producer with its aquaculture output accounting for two-thirds of the global production, plays a decisive role in promoting the development of aquaculture worldwide (Food, Agriculture Organization of the United Nations, 2009). In 2009, China exported 2.94 million metric tons of aquatic products to Japan (19%), the United States (17%), the European Union (17%), Korea (14%), Association of Southeast Asian Nations (12%), Hong Kong (4.3%), Taiwan (2.6%), Russia (2.5%) and other regions (11.6%).

Seafood Network Information Center of China, 2010), were chosen for examining input sources of PHCs to aquaculture environments and potential health risk to global consumers via consumption of farmed fish from this region. The present study extended the findings of previous studies which have documented the occurrence of PHCs such as organochlorine pesticides (OCPs), polychlorinated biphenyls (PCBs) and polybrominated diphenyl ethers (PBDEs) in various environmental compartments of Guangdong Province (e.g., Guan et al., 2007; Guan et al., 2009; Guo et al., 2007a; Guo et al., 2007b;
2.2. Sample preparation and extraction

Gaseous samples were collected using a high-volume air sampler that extracted with 1:1 (v:v) acetone and hexane mixture. Air particle and filtrated with a vermicular system and suspended particulates were filtered with a polyurethane foam plug (PUF) for gases. Air particle samples were also Soxhlet extracted with acetone/hexane mixture. Rain and water samples were collected from two typical mariculture (Hailing Bay and Daya Bay) and freshwater aquaculture zones only. Second, a fugacity-based model of exchange fluxes of PBDEs and DDXs (sum of o,p′- and p,p′-DDT, -DDD, and -DDE and p,p′-DMFU) from environmental media, such as phytoplankton, fish food and water to the fishes via gut and fish gill to determine the predominant routes of exposure to these chemicals. Finally, with the objective of offering some fish consumption advisories for global consumers, the health risk associated with fish consumption was assessed based on the guidelines and methodologies developed by the United States Environmental Protection Agency (USEPA) (U.S. Environmental Protection Agency, 2000).

2. Materials and methods

2.1. Sample collection

Various types of environmental samples, including air, fish (crimson sannper (Lutjanus malabaricus) and snubnose pompano (Trachinotus blochii)), fish food, phytoplankton and water, were collected from two typical mariculture (Hailing Bay and Daya Bay) and two typical freshwater aquaculture (Shunde and Dongguan) zones in South China (Fig. S1) in October 2006 and December 2007. Detailed information on sampling procedures has been reported in our previous studies (Guo et al., 2009b; Zhang et al., 2009, 2010).

2.2. Sample preparation and extraction

In the present study, samples were processed differently depending on the matrix. Fish, fish food, phytoplankton samples were homogenized, freeze-dried, and ground in to fine powder and then Soxhlet extracted with 1:1 (v:v) acetone and hexane mixture. Air particle and gaseous samples were collected using a high-volume air sampler that housed a glass fiber filter for particles and a polyurethane foam plug (PUF) for gases. Air particle samples were also Soxhlet extracted with 1:1 (v:v) acetone and hexane mixture. Rain and water samples, were first filtered with a vermicular system and suspended particulates were collected with glass fiber filters, freeze-dried and Soxhlet extracted. The dissolved organics retained on a glass resin column were eluted and extracted. All the extracts were cleaned with a silica/alumina column. The detailed sample preparation procedures can be found in our previous studies (Guo et al., 2009b; Zhang et al., 2009, 2010).

2.3. Instrument analysis and quality assurance/quality control

Concentrations of DDXs (sum of o,p′- and p,p′-DDT, -DDD, and -DDE and p,p′-DMFU) and PBDEs (sum of BDE-28, -47, -99, -100, -153, -154, -183 and -209) were determined with a Varian 3800 gas chromatograph (GC) interfaced with a Saturn 2000 mass spectrometer (MS) in the selective ion monitoring (SIM) mode and a Shimadzu Model 2010 GC–MS (Shimadzu, Japan) using negative chemical ionization (NCI) in the selected ion monitoring mode. The detailed procedures for the instrumental analysis were described in a previous study (Meng et al., 2007). The quality assurance/quality control results have been reported in our previous studies (Guo et al., 2009b; Yu et al., in press; Zhang et al., 2009, 2010) in press).

2.4. Data analysis

Input fluxes from atmospheric dry and wet depositions, air–water exchange, and diets of fish to the freshwater farming zones have been estimated previously (Zhang et al., 2010, in press). In the present study, the input pathways mentioned above for PHCs as well as emissions of DDXs via antifouling to the mariculture zones were estimated with a similar method. In addition, the probability distributions of input fluxes via various pathways were estimated using Monte Carlo simulation with 5000 trials based on the probability distributions of related parameters (Figs. S2 and S3, using input fluxes of DDXs to the freshwater aquaculture zones through dry deposition as an example and the processes for the other parameters are similar to this). The results are summarized in Table 1 with detailed information presented in the Supplemental Materials.

Furthermore, the major routes of exposure of fishes to PHCs are diffusion across the gill and dietary uptake. Previous studies have predicted rates of accumulation of organic chemicals by fishes via these two routes, and elimination processes occurring by transfer through the gills, in feces, metabolic transformation and growth dilution (Campfens and Mackay, 1997; Catalan and Ventura, 2004; Clark et al., 1990; Gobas, 1993; Mackay, 2001; Mackay and Fraser, 2000). A fugacity-based model of bioaccumulation by fishes, developed by the Canadian Environmental Modeling Center (Mackay and Fraser, 2000), was the primary framework employed in the present study with the aim of estimating the relative importance of each uptake route. Similarly, elimination processes of fishes were investigated by use of both rate constant and fugacity-based simulation models (Eqs. (1) and (2)).

Rate constant format: \[ \frac{dc_i}{dt} = k_i (C_W + C_A - C_I) (K_d + K_a + K_c) \] (1)

Fugacity format: \[ \frac{dF_i}{dt} = D_{WF_i} + C_{DF_i} - F_i (D_W + D_A + D_I) \] (2)

The definitions of the parameters in Eqs. (1) and (2) and descriptions of the model itself can be found in the Supplemental Materials. Some parameters have been modified when used in the present study.

Risks to health of humans associated with fish consumption for global consumers was assessed based on the methods for assessment of risk of both carcinogenic and non-carcinogenic hazard by use of the cancer slope factor (CSF) and reference dose (RfD), respectively developed by the USEPA (U.S. Environmental Protection Agency, 2000) (Eqs. (3) and (4)).

Carcinogenic effects: \[ H_L = \sqrt{\sum_{i=1}^{n} C_{ij} \times CSF_i / BW} \] (3)

Table 1

<table>
<thead>
<tr>
<th>Parameter</th>
<th>Value (mg/m^3)</th>
<th>Value (μg/kg)</th>
<th>Value (g/m^2/yr)</th>
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<tr>
<td>Mean</td>
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<td>4.2 ± 5.9</td>
<td>6500 ± 410</td>
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<tr>
<td>Median</td>
<td>0.22 ± 0.19</td>
<td>2.1 ± 24</td>
<td>190 ± 20</td>
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<tr>
<td>BDE-209</td>
<td>1.3 ± 1.4</td>
<td>5.0 ± 12</td>
<td>80 ± 490</td>
</tr>
<tr>
<td>Marine aquaculture zone</td>
<td></td>
<td></td>
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</tr>
<tr>
<td>DDXs</td>
<td>0.87 ± 0.09</td>
<td>20 ± 18</td>
<td>0.61 ± 0.062</td>
</tr>
<tr>
<td>Marine farmed zone</td>
<td>12 ± 15</td>
<td>5.0 ± 12</td>
<td>6.7 ± 0.66</td>
</tr>
</tbody>
</table>

\(a\) Sum of o,p′- and p,p′-DDT, DDD and DDE and p,p′-DMFU.

\(b\) Sum of BDE-28, -47, -99, -100, -153, -154 and -183.
Noncarcinogenic effects: HQs = ∑

\[ \sum_{j=1}^{n} C_{mj} \times \text{CRR}_j / \text{BW} \times \text{RfD}_m \] (4)

To assess risk of adverse effects on health posed by contaminants, Monte Carlo simulations with five thousand trials were used to produce probability distributions of hazard levels (Fig. S4, using the hazard level based on carcinogenicity of DDXs for Asians as an example and the processes for estimation of hazard levels for the other contaminants are similar to this). Detailed definitions and values for all parameters can be found in the Supplemental Materials. Comparison of values among groups was done using nonparametric tests (Mann–Whitney U and Kruskal–Wallis H) with statistical significance \( p < 0.05 \) under SPSS version 13.0.

3. Results and discussion

3.1. Estimated input fluxes of PHCs to aquaculture zones

Input fluxes of external contaminant sources determine contaminant concentrations in fish in aquaculture. Four major input routes, such as atmospheric wet and dry deposition, air-water exchange, and fish food in the typical aquaculture zones of Guangdong Province, were examined. In addition, current inputs of DDXs via DDT-containing antifouling paint to mariculture zones might also be an important source (Lin et al., 2009). Excluding DDXs for which antifouling paint was the dominant input pathway, input fluxes of PHCs via the diets of fishes in mariculture were one to several orders of magnitude greater than those through other pathways (Daga Bay and Hailing Bay, Fig. S1), whereas input fluxes of DDXs to the freshwater aquaculture zones, such as Shunde and Dongguan (Fig. S1) through diets of fishes were comparable to those via air-water exchange. Input fluxes of PBDE congeners (excluding BDE-209) to aquaculture zones through air-water exchange were also greater compared to other routes. For BDE-209, fluxes via dry and wet depositions accounted for large portions of total BDE-209 input to aquaculture zones, especially to the freshwater aquaculture zones where atmospheric dry and wet depositions were the most dominant input routes. After excluding antifouling paint, fish food was an important source of PHCs to the aquaculture zones (except PBDEs in freshwater aquaculture zones). Fluxes via air-water exchange for DDXs, except for mariculture zones, and PBDE congeners (except BDE-209) in aquaculture zones were also substantial. Dry and wet depositions also appear to be significant for more hydrophobic compounds such as BDE-209. On the other hand, the amount of DDXs via antifouling paint usage is significantly greater than those from other input routes (Table 1).

The results observed in this study are consistent with the conclusion that use of contaminant-free antifouling paint is one of the most effective initiatives for reducing inputs of DDXs to mariculture zones in South China. Another effective means of reducing the inputs of PHCs, especially DDXs is to minimize contaminants in the food fed in aquaculture. Conversely, because PBDEs especially BDE-209, used as ingredients in brominated fire retardants, are mainly derived from primitive handling of electronic waste (e-waste) in China, enhanced law enforcement to restrict the importation and disposal of e-waste in China (Ni et al., 2010) appears to be a necessary step to reduce the loadings of PBDEs in the aquaculture zones of South China.

3.2. Modeled bioaccumulation routes in fish

Once contaminants are transported to aquaculture environments, they are inevitably distributed to various compartments and eventually accumulated by farmed fish. Phytoplankton is an important component in biogeochemical cycling of PHCs in aquatic environments (Russell et al., 1999). In aquaculture zones, PHCs input via various pathways can be accumulated by phytoplankton, which in turn are eaten by fish. Therefore, phytoplankton can also be regarded as one of the food sources for fish. The results of the simulations conducted by use of fugacity-based modeling (Figs. 1 and 2) demonstrated that the relative amounts of contaminants varied among fishes. Mean accumulations of both DDXs and PBDEs via feeding of farmed marine fishes were significantly greater than via diffusion across the gills or via accumulation from phytoplankton (except for BDE-209) (Fig. 1). For farmed freshwater fishes, the mean relative amounts of DDXs via various uptake routes were comparable except for p,p′-DDMU, p,p′-DDD and -DDT for which fish food was the most dominant uptake route. Alternatively, accumulation of all BDE congeners by farmed freshwater fishes via diffusion across the gills was significantly greater than those via other routes (Fig. 2). Therefore, feeding of fish was the major route of exposure for farmed marine fishes to PHCs. Conversely, diffusion across the gills was the predominant exposure route for PBDEs in farmed freshwater fishes, whereas dietary exposure, diffusion across the gills and accumulation via phytoplankton were all significant routes of accumulation of DDXs. This difference is perhaps due to different residual concentrations of target contaminants among fish foods and farming environments. Our previous studies (Gao et al., 2009a; Yu et al., in press; Zhang et al., 2009, in press) reported considerably greater concentrations of target analytes in diets of marine fishes than in diets of freshwater fishes. Also, there were slightly greater contaminant concentrations if freshwater fishes had pond water versus marine aquaculture water. Results of the Monte Carlo simulations confirmed that for most DDX and PBDE congeners, differences in water concentrations contributed the most to differences in fish due to their diet. Concentrations of DDXs and PBDEs in fish food were also an important factor in determining the concentrations of DDXs and PBDEs in farmed marine fishes (Fig. 3).

To estimate net contaminant accumulation in farmed fishes, elimination (loss) processes must be considered. Masses of DDX components eliminated through metabolism were significantly greater than those eliminated via other routes (Figs. S5 and S6), which is consistent with metabolism being an important mechanism for removal of DDXs from both farmed marine and freshwater fishes. Alternatively, removal efficiency for PBDEs was co-congener-specific. That is, the predominant loss route was metabolism for BDE-28, -47 and -209 and growth dilution for BDE-99, -100, -153, -183 and -185 (Fig. 3). The loss mechanism for farmed freshwater fishes was similar to that for farmed marine fishes (Figs. S5 and 6). The sensitivity analysis based on Monte Carlo simulation suggested that half-life time for metabolism in fish (T_M) was a key parameter dictating differences among elimination routes (Fig. 4).

As shown in Equations 539, 543 and 544, if the half-life time in fish approaches the metabolism half-life time (T_M), then the overall loss constant (K loss) would approach the rate constant of metabolic transformation (K_m). In this case, the amounts eliminated through other pathways can be neglected. Therefore, a lesser difference between the half-life times for achieving steady state for the entire exchange process and for metabolism in fish would indicate more predominance of metabolism in the process of elimination and a similar result has been reported by (Clark et al., 1990). In the present study, the metabolism half-life time was assumed to be 900 h for p,p′-DDT, p,p′-DDD and -DDT for which fish food was the dominant uptake route. Alternatively, accumulation of all BDE congeners by farmed freshwater fishes via diffusion across the gills was significantly greater than those via other routes (Fig. 2). Therefore, feeding of fish was the major route of exposure for farmed marine fishes to PHCs. Conversely, diffusion across the gills was the predominant exposure route for PBDEs in farmed freshwater fishes, whereas dietary exposure, diffusion across the gills and accumulation via phytoplankton were all significant routes of accumulation of DDXs. This difference is perhaps due to different residual concentrations of target contaminants among fish foods and farming environments. Our previous studies (Gao et al., 2009a; Yu et al., in press; Zhang et al., 2009, in press) reported considerably greater concentrations of target analytes in diets of marine fishes than in diets of freshwater fishes. Also, there were slightly greater contaminant concentrations if freshwater fishes had pond water versus marine aquaculture water. Results of the Monte Carlo simulations confirmed that for most DDX and PBDE congeners, differences in water concentrations contributed the most to differences in fish due to their diet. Concentrations of DDXs and PBDEs in fish food were also an important factor in determining the concentrations of DDXs and PBDEs in farmed marine fishes (Fig. 3).

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3.3. Potential health risk to global consumers via fish consumption

The risk to health of global consumers exposed to DDXs and PBDEs via consumption of fish exported from Guangdong Province of China was examined. Because estimated concentrations of target chemicals in fish were different for steady and non-steady state conditions, risk to heath of global consumers via consumption of fish farmed in South China was assessed for both steady and non-steady state conditions. Hazard quotients (HQs) for non-carcinogenic effects of DDT and PBDE estimated at the 95% accumulative probability distribution levels (APDLs) under steady state conditions were all less than 1.0 (Table 2). The risks based on carcinogenicity for DDXs and BDE-209 at 95% APDLs were also less than 1.0 × 10⁻³, which is the level of risk recommended by the USEPA (U.S. Environmental Protection Agency, 2000) for global consumers (Table 2). Thus, based on steady-state exposure health risk due to exposure of global consumers to PCBs in fish farmed in China is minimal.

However, the probability of non-steady state health risk based on the non-cancer HQ being greater than 1.0 was 8% for p,p′-DDE and 3% for p,p′-DDT for Africans, 13% for p,p′-DDD, 10% for p,p′-DDE and 10% for p,p′-DDT for Asians, and 12% for p,p′-DDE, 6% for p,p′-DDT and 5% for p,p′-DDT for Europeans. Furthermore, the 95% APDLs for all of the non-steady-state carcinogenic hazards for DDXs were 10-fold greater than the steady-state values. Also, the non-steady-state and steady-state carcinogenic hazard levels are comparable to with PBDEs for global consumers (Table 2). Consumption of fish assumed to have reached steady state exposes consumers to less DDXs compared to fish at non-steady-state. These assumptions are tentative and qualitative because of insufficient toxicological data for PBDEs especially PBDE congeners. The results of the sensitivity analysis by Monte Carlo simulation show that the rate of consumption of fish and proportion of fish in an individual’s diet contribute the most to variation among predicted non-carcinogenic risk due to DDXs and PBDE congeners (Table 57). For carcinogenic hazards, rate of consuming fish in the diet as well as concentrations of p,p′-DDT and p,p′-DDE in food given to farmed marine fishes and PBDE congener in seawater are primary parameters (Table 57). Therefore, different risks due to different fish consumption rates are expected for consumers from different regions (Table 54).

Because PBDEs especially BDE-209 are still used and importation of e-waste has remained active in China (Ni et al., 2010), concentrations of PBDEs in various environmental compartments are expected to continue to increase, resulting in higher human exposure levels via fish consumption. For example, if the annual increase rate is assumed to be 5%, the probability for non-cancer hazard ratios to be higher than unity (currently they are all lower than unity) in 50 years will be 6%, 5%, 14% and 10%, respectively, for Africans, Americans, Asians and Europeans from exposure to BDE-47 via fish consumption.
Because concentrations of PHCs in the wild-caught forage fish are generally greater than those in compound feed (Guo et al., 2009b), use of compound feed manufactured under controlled conditions should be favored in fish farming.

4. Conclusions

Antifouling paint is a predominant source of DDXs to mariculture zones, but its contribution to concentrations in fish has generally been attributed to accumulation via the gills and accumulation through the diet. Atmospheric fluxes are also a significant pathway to the freshwater aquaculture zones for BDE-209 and less brominated BDE congeners (BDE-28, -47, -99, -100, -153, -154 and -183). The results of simulation of accumulation (Figs. 1 and 2) indicate that concentration of PHCs in the food fed to fish is a primary contributor of concentrations of PHCs in fish. Therefore, minimizing the concentrations of PHCs in food fed during farming of fish would be an effective measure for controlling concentrations of PHCs in farmed fishes, especially marine fishes. Consequently, reducing human exposure to these carcinogens can be controlled by regulating concentrations of PHCs in food used in aquaculture. In China, two types of fish food are commonly used; one is compound feed made of fish powders, fish oil, flour, wheat protein powders and soybean meal and the other one is a collection of small wild fish caught in the deep sea (Food, Agriculture Organization of the United Nations, 2007). Because concentrations of PHCs in the wild-caught forage fish are generally greater than those in compound feed

Table 2

Non-cancer (hazard quotient (HQ)) of human exposure to non-carcinogenic exposure limit-reference dose (RID) and cancer hazard (exposure level multiplied by upper bond of the lifetime cancer risk-cancer slope factor (CSF)) values at 95% accumulative probability distribution for global consumers. Bold numbers are non-cancer HQs and cancer hazard levels that are greater than the assessment criteria of unity and 1.0 × 10⁻⁴, respectively.

<table>
<thead>
<tr>
<th></th>
<th>Africa</th>
<th>America</th>
<th>Asia</th>
<th>Europe</th>
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<td><strong>Non-cancer HQs</strong></td>
<td></td>
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<tr>
<td>p,p'-DDD</td>
<td>0.062</td>
<td>0.084</td>
<td>0.20</td>
<td>0.12</td>
<td>0.053</td>
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<td>p,p'-DDE</td>
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<td>0.55</td>
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<tr>
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<td>BDE-209</td>
<td>3.1 × 10⁻¹⁰</td>
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Cancer hazard level

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<th>DDX(10⁻⁴)</th>
<th>DDX(10⁻⁴)</th>
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<td></td>
<td>2.0 × 10⁻⁵</td>
<td>1.8 × 10⁻⁵</td>
<td>3.5 × 10⁻⁵</td>
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<tr>
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<td>1.2 × 10⁻¹⁰</td>
<td>1.1 × 10⁻¹⁰</td>
<td>1.7 × 10⁻¹⁰</td>
<td>3.6 × 10⁻¹⁰</td>
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* Sum of p,p'- and p,p'-DDE, DDD and DDE and p,p'-DDMU.

Acknowledgements

This work was financially supported by the National Natural Science Foundation of China (Nos. 40821003 and U0633005) and the Chinese Academy of Sciences (KZCX2-YW-Q02-06-01). We are also grateful to Ru-Lang Shen, Ying Guo, Zhi-Qing Shan, Yi-Yi Yu, Qiu-Xin Huang, and Xiang-Fei Sun for sample collection and Lian-Jun Bao for laboratory support. Prof. Giesy was supported by the Canada Research Chair program, an at large Chair Professorship at the Department of Biology and Chemistry and State Key Laboratory in Marine Pollution, City University of Hong Kong, The Einstein Professor Program of the Chinese Academy of Sciences and the Visiting Professor Program of King Saud University. This is contribution No. IS-1336 from GACCS.

Appendix A. Supplementary data

Supplementary data to this article can be found online at doi:10.1016/j.envint.2011.04.012.
H.-Y. Yu et al. / Environment International 37 (2011) 1190–1195

1195

References


Supplemental Materials

MANUSCRIPT TITLE: Persistent halogenated compounds in aquaculture environments of South China: Implications for global consumers’ health risk via fish consumption

AUTHORS: Huan-Yun Yu, Bao-Zhong Zhang, John P. Giesy, and Eddy Y. Zeng

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NO. OF FIGURES: 6
NO. OF TABLES: 7
NO. OF PAGES: 44
Procedures for estimating input fluxes

The input fluxes from atmospheric dry deposition ($F_{\text{dry}}$), wet deposition ($F_{\text{wet}}$), air-water exchange ($F_{\text{gas}}$), fish feeding ($F_{\text{feed}}$) and antifouling discharge ($F_{\text{anti}}$) to the aquaculture zones are estimated with the following equations:

\[ F_{\text{dry}} = C_{\text{particle}} V_d \]  \hspace{1cm} (S1)

\[ F_{\text{wet}} = C_{\text{rain}} P \]  \hspace{1cm} (S2)

\[ F_{\text{gas}} = K_g (C_{\text{gas}} - C_{\text{water}} H/RT) \]  \hspace{1cm} (S3)

\[ F_{\text{feed}} = C_{\text{feed}} m_r \]  \hspace{1cm} (S4)

\[ F_{\text{anti}} = I_{\text{input}}/A \]  \hspace{1cm} (S5)

where

- $A$: Area of the aquaculture zone under investigation in the present study (703 km$^2$)

(51766.com 2010)

- $C_{\text{particle}}$: Concentration measured in the air particle phase ($\mu$g/m$^3$)

- $C_{\text{water}}$: Concentration in filtered fishpond water ($\mu$g/m$^3$)

- $C_{\text{gas}}$: Concentration in the air gaseous phase of air ($\mu$g/m$^3$)

- $C_{\text{feed}}$: Concentration in fish feed ($\mu$g/kg wet wt)

- $C_{\text{rain}}$: Concentration in rain ($\mu$g/m$^3$)

- $H$: Henry’s Law constant (Pa m$^3$/mol)

- $I_{\text{input}}$: Antifouling input amount (~12–24 tons/yr, accounting for 2/5 of the total in Guangdong Province (Wang et al. 2007))

- $K_g$: Overall gas-phase mass transfer coefficient (m/yr)
$m_r$: Mass input rate via feeding to fishpond (kg dry wt/m²-yr)

$P$: Precipitation rate (m/yr)

$R$: Ideal gas constant (8.31 Pa m³/K mol)

$T$: Absolute temperature (K)

$V_d$: Particle depositional velocity in air (m/yr)

Definitions of all parameters in the fugacity-based fish bioaccumulation modeling are as follows (Gobas 1993; Clark et al. 1990):

$C_{A1F}$: Chemical concentration in phytoplankton (mol/m³)

$C_{A2F}$: Chemical concentration in fish food (mol/m³)

$C_F$: Chemical concentration in fish body (mol/m³)

$C_F$: Chemical concentration in the water suspend particulate phase (g/kg)

$C_{Phy}$: Phytoplankton concentration in water (kg/L)

$C_W$: Chemical concentration in the water dissolved phase (g/m³)

$C_{total-W}$: Chemical concentration in water (dissolved and particulate phases) (g/m³)

$C_{particle}$: Concentration of suspended particles in water (g/m³)

$D_E$: Transfer parameter for chemical elimination in the faeces (mol/Pa·h)

$D_G$: Transfer parameter for growth dilution (mol/Pa·h)

$D_M$: Transfer parameter for metabolic transformation of the chemical in fish (mol/Pa·h)

$D_W$: Transfer parameter between fish and water across gills (mol/Pa·h)

$E_A$: Food uptake efficiency

$f_F$: Chemical’s fugacity in fish (Pa)

$Flux_{A}$: Uptake flux from food (ng/day)
Flux_E: Loss flux via egestion (ng/day)

Flux_G: Loss flux via growth dilution (ng/day)

Flux_M: Loss flux via metabolism (ng/day)

Flux_phy: Uptake flux from phytoplankton (ng/day)

Flux_W: Uptake flux via gills (ng/day)

Flux_2: Loss flux via gills (ng/day)

f_w: Chemical’s fugacity in water (Pa)

G_A: Feeding rate (kg/day)

H: Henry’s Law constant (Pa.m/mol)

K_A: Rate constant of chemical uptake from fish food (1/h)

K_W: Rate constant of chemical uptake from water (1/h)

K_W': Rate constant of chemical loss via gills (1/h)

K_E: Rate constant of elimination by egestion (1/h)

K_FW: Partition coefficient between water and fish

K_G: Rate constant of growth dilution (1/h)

K_M: Rate constant of metabolic transformation (1/h)

K_OW: Octanol-water partition coefficient

K_total: Overall constant for loss (1/h)

L_A1F: Lipid content in phytoplankton

L_A2F: Lipid content in fish feed

L_F: Lipid content in fish

M: Molar mass of the chemical (g/mol)
$P_1$: The fraction of fish diet that is consisted of phytoplankton

$P_{chl-a}$: Chlorophyll-$a$ concentration in mariculture zones (Hailing Bay and Daya Bay) (kg/L)

$\rho_{phy}$: The density of phytoplankton ($1.0 \times 10^3$ kg/m$^3$)

$Q$: Digestion coefficient ($Q = 3$)

$R_{TO}$: Gill membrane organic phase resistance (h)

$R_W$: Aqueous gill resistance (h)

$t_{M1/2}$: Chemical metabolism half-life time in fish

$t_{S1/2}$: Overall half-time to steady state (h)

$V_F$: Fish volume (m$^3$)

$W_F$: Fish weight (kg)

$y$: Organic carbon content in the water particulate phase ($y = 0.04$)

$Z_A$: Chemical’s fugacity capacity in fish food (mol/m·Pa)

$Z_O$: Chemical’s fugacity capacity in octanol (mol/m·Pa)

$Z_W$: Chemical’s fugacity capacity in water (mol/m·Pa)

Treating the fish as one entity, the entire exchange fluxes between fish body and water and food can be described by
The equations used in the model (Gobas 1993; Clark et al. 1990; Arnot and Gobas 2004; Mackay 2001):

The uptake flux from water \( (Flux_w) \) can be estimated with Equations S6-S15.
\[ Flux_W = 24 \times 10^9 D_W f_w M \]  \hspace{1cm} (S6)

\[ f_w = C_W/Z_W \]  \hspace{1cm} (S7)

\[ Z_W = 1/H \]  \hspace{1cm} (S8)

\[ C_{total-W} = 10^{-3} C_P C_{particle} + C_W \]  \hspace{1cm} (S9)

\[ 10^3 C_P / C_W = 0.41yK_{OW} \ (y = 0.04) \]  \hspace{1cm} (S10)

\[ D_W = K_W V_F Z_W \]  \hspace{1cm} (S11)

\[ K_W = K_W' L_f K_{OW} \]  \hspace{1cm} (S12)

\[ K_W' = 1/(R_W K_{OW} + R_{TO}) \]  \hspace{1cm} (S13)

\[ R_W = 0.15 V_F^{0.36} \]  \hspace{1cm} (S14)

\[ R_{TO} = 12600 V_F^{0.29} \]  \hspace{1cm} (S15)

The uptake flux from fish food (Flux\textsubscript{A}) can be estimated with Equations S16-S18

\[ Flux_A = 10^9 E_A (G_A - G_v C_{phy}) C_{A1F} M V_F / W_F \]  \hspace{1cm} (S16)

\[ E_A = 1/(3 \times 10^{-7} K_{OW} + 2) \]  \hspace{1cm} (S17)

\[ G_A = 0.022 \exp(0.067 W_F^{0.85}) \]  \hspace{1cm} (S18)

The uptake flux from phytoplankton uptake (Flux\textsubscript{phy}) can be estimated with Equations S19-S20

\[ Flux_{phy} = 10^9 E_A G_v C_{phy} C_{A1F} M V_F / W_F \]  \hspace{1cm} (S19)

\[ G_v = 1400 W_F^{0.65}/(-0.24T + 14.04)/0.9 \]  \hspace{1cm} (S20)

Chemical concentration in phytoplankton (C\textsubscript{A1F}) of freshwater fish pond water was estimated by Equations S21-S23 due to no phytoplankton samples collected (Meylan et al. 1999)

\[ \text{Bioconcentration factor (BCF)} = 10^3 C_{A1F} M / \rho_{phy}/C_w \]  \hspace{1cm} (S21)

\[ \log \text{BCF} = 0.77 \log K_{OW} - 0.70 + 0.62 \log K_{OW} \text{ 1 to 7} \]
\[ \log \text{BCF} = 1.37 \log \text{K}_{\text{OW}} + 14.4 + 0.62 \log \text{K}_{\text{OW}} > 7 \]  

(S22)

The phytoplankton concentration \( C_{\text{phy}} \) in mariculture zones can be estimated with Equation S24 and the values for \( P_{\text{chl-a}} \) were determined according to previous studies (Qiu et al. 2006)

\[ P_{\text{chl-a}} = 1.12 C_{\text{phy}} + 0.21 \]  

(S24)

The phytoplankton concentration \( C_{\text{phy}} \) in freshwater aquaculture zones was 20 mg/L (Wang et al. 1994)

The loss flux via gills (\( \text{Flux}_2 \)) can be estimated with Equations S25-S27

\[ \text{Flux}_2 = 24 \times 10^9 D_{\text{WF}} f_M \]  

(S25)

\[ K_{\text{FW}} = Z_F/Z_W = L_F \text{K}_{\text{OW}} \]  

(S26)

\[ f_F = (\text{Flux}_W + \text{Flux}_A)/M/24/10^9/(D_W + D_E + D_M + D_G) \]  

(S27)

The loss flux via egestion (\( \text{Flux}_E \)) can be estimated with Equations S28-S36

\[ \text{Flux}_E = 24 \times 10^9 (D_{\text{E1F}} + D_{\text{E2F}}) f_E M \]  

(S28)

\[ D_{\text{E1F}} = D_{\text{A1F}}/Q \]  

(S29)

\[ D_{\text{E2F}} = D_{\text{A2F}}/Q \]  

(S30)

\[ D_{\text{A1F}} = V_{\text{F}} K_{\text{A1F}} Z_{\text{A1F}} \]  

(S31)

\[ D_{\text{A2F}} = V_{\text{F}} K_{\text{A2F}} Z_{\text{A2F}} \]  

(S32)

\[ Z_{\text{A1F}} = L_{\text{A1F}} Z_O = L_{\text{A1F}} \text{K}_{\text{OW}} /H \]  

(S33)

\[ Z_{\text{A2F}} = L_{\text{A2F}} Z_O = L_{\text{A2F}} \text{K}_{\text{OW}} /H \]  

(S34)

\[ K_{\text{A1F}} = G_v C_{\text{phy}} E_A/W_F/24 \]  

(S35)

\[ K_{\text{A2F}} = (G_A-G_v C_{\text{phy}}) E_A/W_F/24 \]  

(S36)
The loss flux via metabolism ($\text{Flux}_M$) can be estimated with Equations S37-S39

\[
\text{Flux}_M = 24 \times 10^9 D_M f_M M
\] (S37)

\[
D_M = K_M V_F Z_F
\] (S38)

\[
K_M = 0.693/t_M^{1/2}
\] (S39)

The loss flux via growth dilution ($\text{Flux}_G$) can be estimated with Equations S40-S42

\[
\text{Flux}_G = 24 \times 10^9 D_G f_M M
\] (S40)

\[
D_G = K_G V_F Z_F
\] (S41)

\[
K_G = 0.00251 V_F^{-0.2}/24
\] (S42)

Half-time to achieve steady state for the entire exchange process is estimated with Equations S43-S46

\[
t_{S1/2} = 0.693/K_{total}
\] (S43)

\[
K_{total} = K_M + K_G + K_E + K_{W'}
\] (S44)

\[
K_E = D_E/V_F Z_F
\] (S45)

\[
D_E = D_{E1F} + D_{E2F}
\] (S46)

Combining Equations S6-S24 which describes the relative uptake amounts via fish food in fish ($P_{uptake}$) yields:

\[
P_{uptake} = V_F (0.022 e^{0.06T} W_F^{0.85} - 1400 W_F^{0.65}/(-0.24T + 14.04)/0.9
\]

\[
\times C_{phy})MC_{A2F}/W_F/(3 \times 10^{-7} K_{OW} + 2)/(24 V_F K_{OW} L_C W/(0.15 V_F^{0.36} K_{OW} + 12600 V_F^{0.29})
\plus V_F (0.022 e^{0.06T} W_F^{0.85} - 1400 W_F^{0.65}/(-0.24T + 14.04)/0.9 C_{phy})MC_{A2F}/W_F/(3 \times
\times 10^{-7} K_{OW} + 2) + 1400 W_F^{0.65}/(-0.24T + 14.04)/0.9 C_{phy}MC_{A1F} V_F/W_F/(3 \times 10^{-7} K_{OW} + 2))
\] (S47)
Combining equations S25-S46, equations S48-S51 which describe the relative losses in fish can \( (P_{\text{loss}}) \) yields:

\[
P_{\text{loss via gill}} = \frac{24}{(0.15V_F^{0.36}K_{OW} + 12600V_F^{0.29})}/(24/(0.15V_F^{0.36}K_{OW} + 12600V_F^{0.29}) + 16.6/t_M^{1/2} + 0.00251V_F^{-0.2} + (0.022e^{0.06T}W_F^{0.85} - 1400W_F^{0.65}/(-0.24T + 14.04)/0.9C_{\text{phy}}L_{A2F}/W_F/L_F/Q/(3 \times 10^{-7}K_{OW} + 2) + 1400W_F^{0.65}/(-0.24T + 14.04)/0.9C_{\text{phy}}L_{A1F}/W_F/L_F/Q/(3 \times 10^{-7}K_{OW} + 2)) \tag{S48}
\]

\[
P_{\text{loss via metabolism}} = \frac{16.6/t_M^{1/2}}{(24/(0.15V_F^{0.36}K_{OW} + 12600V_F^{0.29}) + 16.6/t_M^{1/2} + 0.00251V_F^{-0.2} + (0.022e^{0.06T}W_F^{0.85} - 1400W_F^{0.65}/(-0.24T + 14.04)/0.9C_{\text{phy}}L_{A2F}/W_F/L_F/Q/(3 \times 10^{-7}K_{OW} + 2) + 1400W_F^{0.65}/(-0.24T + 14.04)/0.9C_{\text{phy}}L_{A1F}/W_F/L_F/Q/(3 \times 10^{-7}K_{OW} + 2)) \tag{S49}
\]

\[
P_{\text{loss via growth}} = 0.00251V_F^{-0.2}/(24/(0.15V_F^{0.36}K_{OW} + 12600V_F^{0.29}) + 16.6/t_M^{1/2} + 0.00251V_F^{-0.2} + (0.022e^{0.06T}W_F^{0.85} - 1400W_F^{0.65}/(-0.24T + 14.04)/0.9C_{\text{phy}}L_{A2F}/W_F/L_F/Q/(3 \times 10^{-7}K_{OW} + 2) + 1400W_F^{0.65}/(-0.24T + 14.04)/0.9C_{\text{phy}}L_{A1F}/W_F/L_F/Q/(3 \times 10^{-7}K_{OW} + 2)) \tag{S50}
\]

\[
P_{\text{loss via egestion}} = \frac{((0.022e^{0.06T}W_F^{0.85} - 1400W_F^{0.65}/(-0.24T + 14.04)/0.9C_{\text{phy}}L_{A2F}/W_F/L_F/Q/(3 \times 10^{-7}K_{OW} + 2) + 1400W_F^{0.65}/(-0.24T + 14.04)/0.9C_{\text{phy}}L_{A1F}/W_F/L_F/Q/(3 \times 10^{-7}K_{OW} + 2))}{(24/(0.15V_F^{0.36}K_{OW} + 12600V_F^{0.29}) + 16.6/t_M^{1/2} + 0.00251V_F^{-0.2} + (0.022e^{0.06T}W_F^{0.85} - 1400W_F^{0.65}/(-0.24T + 14.04)/0.9C_{\text{phy}}L_{A2F}/W_F/L_F/Q/(3 \times 10^{-7}K_{OW} + 2) + 1400W_F^{0.65}/(-0.24T + 14.04)/0.9C_{\text{phy}}L_{A1F}/W_F/L_F/Q/(3 \times 10^{-7}K_{OW} + 2))} \tag{S51}
\]

Concentration of target analytes in fish body at steady state \( (C_{F-\text{steady}}) \) can be estimated by:
\[ C_{F,\text{steady}} = (L_F K_{\text{OW}} C_W/(0.15 V_F^{0.36} K_{\text{OW}} + 12600 V_F^{0.29}) + (0.022 e^{0.06 T} W_F^{0.85} -1400 W_F^{0.65}/(-0.24 T + 14.04)/0.9 C_{\text{phy}} M C_{A2F}/24/W_F/(3 \times 10^{-7} K_{\text{OW}} + 2) +1400 W_F^{0.65}/0.9/(-0.24 T + 14.04)/(3 \times 10^{-7} K_{\text{OW}} + 2)C_{\text{phy}} M C_{A1F}/W_F/24)/ (1/(0.15 V_F^{0.36} K_{\text{OW}} + 12600 V_F^{0.29}) + 0.693/t_{\text{M1/2}} + (0.022 e^{0.06 T} W_F^{0.85} -1400 W_F^{0.65}/(-0.24 T + 14.04)/0.9 C_{\text{phy}} L_{A2F}/24/W_F/(3 \times 10^{-7} K_{\text{OW}} + 2)/Q/L_F + 1400 W_F^{0.65}/0.9/(-0.24 T + 14.04)C_{\text{phy}}/(3 \times 10^{-7} K_{\text{OW}} + 2)L_{A1F}/24/W_F/Q/L_F + 0.00251 V_F^{-0.2}/24)\] (S52)

The detailed definitions for all parameters of health risk assessment are as follows:

**BW** = Consumer body weight (kg)

**C_{mj}** = Concentration of chemical contaminant \( m \) in fish species \( j \) (mg/kg)

**CR_{j}** = Daily consumption rate of fish species \( j \) (kg/d)

**CSF** = Cancer slope factor, usually the upper 95 percent confidence limit on the linear term in the multistage model used by the USEPA 1/(mg/kg·d)

**HQs** = Hazard quotients

**HLs** = Hazard levels

**P** = Proportion of fish in an individual’s diet

**RfD** = Reference dose (mg/kg·d)
Table S1

Physical and chemical properties of the target analytes under investigation.

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<tr>
<th>Chemical</th>
<th>$M$ (g/mol)$^a$</th>
<th>$\log K_{ow}$ $^a$</th>
<th>Half –life time (h)$^b$</th>
<th>$H$ (Pa.m$^3$/mol)$^a$</th>
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<td>$o,o'$-DDD</td>
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<td>6.23</td>
<td>360</td>
<td>0.35</td>
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<tr>
<td>$p,p'$-DDD</td>
<td>320.04</td>
<td>6.5</td>
<td>360</td>
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</tr>
<tr>
<td>$o,o'$-DDT</td>
<td>354.5</td>
<td>6.76</td>
<td>900</td>
<td>1.31</td>
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<tr>
<td>$p,p'$-DDT</td>
<td>354.5</td>
<td>6.91</td>
<td>900</td>
<td>1.31</td>
</tr>
<tr>
<td>$o,o'$-DDE</td>
<td>318.04</td>
<td>6.94</td>
<td>360</td>
<td>7.95</td>
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<td>$p,p'$-DDE</td>
<td>318.04</td>
<td>6.96</td>
<td>360</td>
<td>7.95</td>
</tr>
<tr>
<td>$M$ (g/mol)</td>
<td>$\log K_{ow}$ $^c$</td>
<td>Half –life time (h)$^d$</td>
<td>$H$ (Pa.m$^3$/mol)$^e$</td>
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</tr>
<tr>
<td>BDE-47</td>
<td>485.8</td>
<td>6.81</td>
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<td>1.5</td>
</tr>
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<td>BDE-99</td>
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<td>5000</td>
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</tr>
<tr>
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<td>0.069</td>
</tr>
<tr>
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</tr>
<tr>
<td>BDE-154</td>
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<td>1000</td>
<td>0.09</td>
</tr>
</tbody>
</table>

$^a$ Refer to (Kelly et al. 2007).

$^b$ Refer to EPI Suite model BIOWIN 4 for water half–life time.

$^c$ Refer to (Braekevelt et al. 2003).

$^d$ Refer to (Bhavsar et al. 2008).

$^e$ Refer to (Xu et al. 2007; Tittlemier et al. 2002).
Table S2

Properties of farmed marine fishes (snubnose pompano (SP) and crimson snapper (CS)) and sea water collected from Hailing Bay (HLB) and Daya Bay (DYB) of South China.

<table>
<thead>
<tr>
<th>Sample name</th>
<th>$V_F$</th>
<th>$W_F$</th>
<th>$L_F$</th>
<th>$L_{A2F}$</th>
<th>$L_{A1F}$</th>
<th>$C_S^c$</th>
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<tbody>
<tr>
<td>HLB-1⁰b-SPc-28d</td>
<td>0.53</td>
<td>0.50</td>
<td>0.12</td>
<td>0.078</td>
<td>0.066</td>
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</tr>
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<td>HLB-1-SP-30</td>
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<td>0.54</td>
<td>0.14</td>
<td>0.078</td>
<td>0.066</td>
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</tr>
<tr>
<td>HLB-2-SP</td>
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<td>0.10</td>
<td>0.06</td>
<td>0.078</td>
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<td>HLB-3-SP-5</td>
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<td>0.41</td>
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</tr>
<tr>
<td>HLB-5-SP-52</td>
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</tr>
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</tr>
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</tr>
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<tr>
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<tr>
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</tr>
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<td>Value4</td>
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<tr>
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<tr>
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</tr>
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<td>0.12</td>
<td>0.039</td>
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</tr>
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<td>Fish Name</td>
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<td>a</td>
<td>b</td>
<td>c</td>
<td>d</td>
<td>e</td>
</tr>
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<td>0.039</td>
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<td>DYB-1,2-CS-22</td>
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</tr>
<tr>
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<td>0.44</td>
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<td>0.059</td>
<td>0.039</td>
<td>0.066</td>
<td>54.8</td>
</tr>
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<td>DYB-1,2-CS-24</td>
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<td>0.078</td>
<td>0.039</td>
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</tr>
<tr>
<td>DYB-4-CS-1</td>
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<td>0.18</td>
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</tr>
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<td>0.039</td>
<td>0.066</td>
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</tr>
<tr>
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<td>0.039</td>
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<td>0.039</td>
<td>0.066</td>
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</tr>
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<td>0.039</td>
<td>0.066</td>
<td>26.7</td>
</tr>
</tbody>
</table>

\[ a \text{ Sampling location name.} \]
\[ b \text{ Sampling location code.} \]
\[ c \text{ Fish name.} \]
\[ d \text{ Fish sample code.} \]
\[ e \text{ Suspend particular concentration in water (g/m}^3)\text{.} \]
Table S3

Properties of farmed freshwater fishes collected from two typical freshwater cultured fish ponds of Shunde (SD) and Dongguan (DG), South China.

<table>
<thead>
<tr>
<th></th>
<th>$L_F$</th>
<th>$V_F$</th>
<th>$L_{A2}$</th>
<th>$W_F$</th>
</tr>
</thead>
<tbody>
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<td>DG-F-1-CY$^f$-1</td>
<td>0.11</td>
<td>0.88</td>
<td>0.037</td>
<td>0.84</td>
</tr>
<tr>
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<td>0.55</td>
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<td>0.53</td>
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<td>DG-F-1-YY$^g$-1</td>
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<td>0.89</td>
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<td>0.20</td>
<td>0.037</td>
<td>0.19</td>
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<td>DG-F-3-TTF$^h$-2</td>
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<td>0.43</td>
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<td>0.46</td>
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<td>0.037</td>
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<td>0.073</td>
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<td>0.46</td>
<td>0.073</td>
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<tr>
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<tr>
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<td>Sampling location name</td>
<td>Farmed freshwater fish</td>
<td>Sampling location code</td>
<td>Fish name</td>
<td>Fish sample code</td>
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\(^a\) Sampling location name.

\(^b\) Farmed freshwater fish.

\(^c\) Sampling location code.

\(^d\) Fish name- Tilapia (*Tilapia*).

\(^e\) Fish sample code.

\(^f\) Grass carp (*Ctenopharyngodon idellus*).
g Bighead carp (*Aristichthys nobilis*).

h Bluntsnout bream (*Magalobrama amblycephala*).

i Crucian carp (*Carassius auratus*).

j Common mullet (*Mugilcephalus*).

k Largemouth bass (*Micropterus salmoides*).

l Mud carp (*Cirrhinus molitorella*).

m Common carp (*Cyprinus carpio*).

The values for $L_{A1F}$ are the same as those in Table S2
### Table S4
Consumption rates of farmed marine and freshwater fish (g/day) for global consumers (Food and Agriculture Organization of the United Nations 2007).

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**Oceania**

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Table S5
Proportion of fish (sum of farmed marine and freshwater fish) in an individual’s diet for global consumers
(Food and Agriculture Organization of the United Nations 2007).

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Table S6
Risk values for DDX and PBDE congeners.

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<th>CSF (per mg/kg·day)</th>
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\(^a\) The values of RfD and CSF are assumed the same as those of \( p,p'-\text{DDT} \).

\(^b\) The value of RfD is assumed the same as that of BDE-99.
Table S7
Results of sensitivity analysis for contribution of main parameters to the variances of health risk levels for global consumers exposed to DDX and PBDE congeners via consumption of fish exported from South China based on Equations S52, 3and 4 using Monte Carlo simulation.

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### Noncancer_BDE-209

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### Lifetime risk_DDXs

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### Lifetime risk_BDE-209

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a Farmed marine fish consumption rate for African consumers (kg/day).

b Proportion of fish in an individual’s diet.

c Concentration level in seawater (g/m³).

d Concentration level in marine fish feed (g/m³).

e Farmed freshwater fish consumption rate for Asian people (kg/day).

f Farmed marine fish lipid content.

g Concentration level in freshwater cultured fish pond water (g/m³).

h Farmed freshwater fish lipid content.
Figure S1. Map of the study areas in South China and the locations of two estuarine bay (Hailing Bay and Daya Bay) and four freshwater cultured fish ponds (two each for Shunde and Dongguan, respectively) where sampling was conducted.
Figure S2. Concentration of DDXs (sum of o,p'- and p, p'-DDT, DDD and DDE and p,p'-DDMU) in air particulate collected from the freshwater aquaculture zones fit lognormal distributions.
Figure S3. Input fluxes of DDXs (sum of \(o,p'-\) and \(p,p'-\)DDT, DDD and DDE and \(p,p'-\)DDMU) to the freshwater aquaculture zones through dry depositions using Monte Carlo simulation with 5000 trials fit lognormal distributions and the median value is 1.1 \(\mu g/m^2\) yr.
Figure S4. The hazard level based on carcinogenicity of DDXs (sum of $o,p'$- and $p$, $p'$-DDT, DDD and DDE and $p,p'$-DDMU) for Asians at 95% accumulative probability distribution levels is $3.4 \times 10^{-5}$ risk.
Figure S5. Relative losses of DDXs accounting for total loss in (a) farmed marine fish and (b) farmed freshwater fish.
Figure S6. Relative losses of PBDEs accounting for total loss in (a) famed marine fish and (b) farmed freshwater fish.
References


