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

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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, 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 -DDT) 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.

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). Pertinent to the Stockholm Convention on persistent organic pollutants (POPs) (Stockholm Convention on POPs), human exposure to POPs has been subject to rigorous examination. Consumption of aquatic products is a major route of exposure of humans to persistent halogenated compounds (PHCs), a main subset of POPs, and an important source of health risk (Yu et al., 2010). Therefore, the quality of aquatic products from China bears substantial implications for global consumers' health safety.

To effectively limit concentrations of residues in fishes in culture, it is necessary to know the pathways of exposure. Therefore, the current study was conducted to determine the relative magnitudes of accumulation of several residues by fishes during aquaculture. In the present study, four typical aquaculture zones (two each for marine and freshwater aquaculture, respectively) in Guangdong Province of South China (Fig. S1 of the Supplemental Materials; "S" indicates tables and figures in the Supplemental Materials thereafter), which is the second-largest aquaculture base in China accounting for 13.5% of total aquaculture exports by China in 2009 (Liu, 2008; 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;

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Guo et al., 2009a; Mai et al., 2005; Meng et al., 2007; Wu et al., 2008; Zhang et al., 2009). In particular, concentrations of PHCs in typical fish farming zones of Guangdong Province were determined (Guo et al., 2009b; Yu et al., in press; Zhang et al., 2010, in press). Based on the results of previous studies, it had been speculated that occurrence of PHCs in farmed fish might be derived mainly from fish food (Guo et al., 2009b; Hites et al., 2004; Minh et al., 2006).

The present study was multi-tiered. First, source diagnostics were conducted via estimation of various input fluxes to the farming environments, including atmospheric dry and wet depositions, air–water exchange, fish feeding and releases from use of antifouling paint (for marine aquaculture zones only). Second, a fugacity-based model of accumulation by fishes was used to quantitatively evaluate exchange fluxes of PBDEs and DDXs (sum of *o,p′*- and *p,p′*-DDT, -DDD, and -DDE and *p,p′*-DDMU) 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, rain, fish (crimson sanpper (*Lutjanus malabaricus*) and snubnose pompano (*Trachinotous 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′*-DDMU) 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 fish 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 fish were investigated by use of both rate constant and fugacity-based simulation models (Eqs. (1) and (2)):

$$\text{Rate constant format}: dC_F/dt = K_1C_W + K_AC_A - C_F(K_2 + K_M + K_E + K_G) \tag{1}$$

$$\text{Fugacity format}: dV_FZ_Ff_F/dt = D_Wf_W + C_Af_A - f_F(D_W + D_E + D_M + D_G) \tag{2}$$

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))

$$\text{Carcinogenic effects}: HLs = \left(\sum_{j=1}^{n} \mathrm{C}_{mj} \times \mathrm{CR}_j\right) \times \mathrm{CSF_m/BW} \tag{3}$$

Table 1

Input fluxes (median ± standard deviation; in μg/m^{2}yr) via atmospheric dry deposition (F_{dry}), wet deposition (F_{wet}), air–water exchange (F_{gas}), fish food (F_{feed}) and antifouling paint discharge (F_{anti}).

	F_{dry}	F_{wet}	F_{gas}	F_{feed}	F_{anti}
Marine aquaculture zone					
DDXs[a]	0.53 ± 1.0	0.80 ± 0.09	4.2 ± 5.9	6500 ± 810	17,000–34,000
Σ_7PBDE[b]	0.22 ± 1.0	0.34 ± 0.19	2.1 ± 24	190 ± 20	
BDE-209	1.3 ± 14	5.0 ± 12	0.14 ± 0.88	80 ± 490	
Freshwater farming zone					
DDXs[a]	1.1 ± 1.3	0.80 ± 0.09	17 ± 29	12 ± 1.2	
Σ_7PBDE[b]	0.87 ± 0.37	0.34 ± 0.19	2.0 ± 18	0.61 ± 0.062	
BDE-209	12 ± 15	5.0 ± 12	6.4 ± 3.0	6.7 ± 0.66	

[a] Sum of *o,p′*- and *p,p′*-DDT, DDD and DDE and *p,p′*-DDMU.

[b] Sum of BDE-28, -47, -99. -100. -153, -154 and -183.

$$\text{Noncarcinogenic effects}: HQs = \left(\sum_{j=1}^{n} \mathrm{C}_{mj} \times \mathrm{CR}_{j}\right)/\mathrm{BW}/\mathrm{P}/\mathrm{RfD}_{m} \quad (4)$$

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 (Daya 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, o,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 (Guo 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 fish 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).

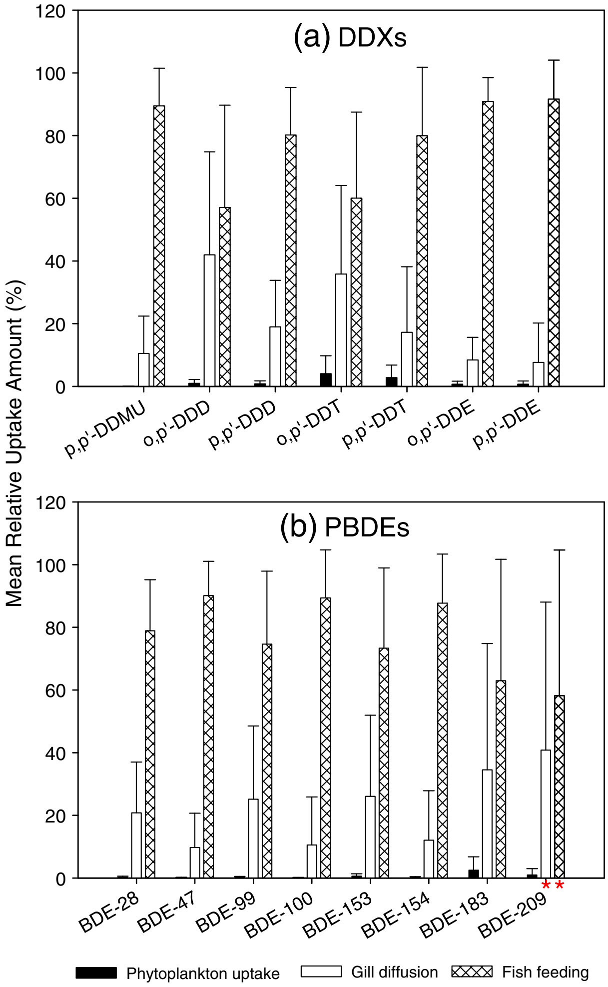

Fig. 1. Mean relative accumulations of DDXs through consumption of phytoplankton, diffusion across gills and through the diet (fish food) for (a) DDXs and (b) PBDEs in farmed marine fishes. The bars with symbol * above the x coordinate indicate no significant difference ($p>0.05$) between the two relative uptake amounts for BDE-209.

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 congener-specific. That is, the predominant loss route was metabolism for BDE-28, -47 and -209 and growth dilution for BDE-99, -100, -153, -154 and -183 (Fig. S6). 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 S39, S43 and S44, if the half-life time ($t_{S1/2}$) for achieving steady state in fish approaches the metabolism half-life time ($t_{M1/2}$), then the overall loss constant (K_{total}) 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 o,p'-DDT and p,p'-DDT, 360 h for other DDT components, 1000 h for BDE-28 and -47, 5000 h for BDE-99 and BDE-100, 6000 h for BDE-153 and BDE-154, 2500 h for BDE-183 and 1000 h for BDE-209, respectively. The mean half-times to reach steady state in farmed marine fishes calculated (Equations S13–S15, S29–S36, S39 and S42–S46 in the Supporting Information) were 225, 244, 251, 457, 470, 264, 264, 416, 483, 950, 929, 1101, 1089, 898 and 591 h for p,p'-DDMU, o,p'-DDD, p,p'-DDD, o,p'-DDT, p,p'-DDT, o,p'-DDE, p,p'-DDE, BDE-28, BDE-47, BDE-99, BDE-100, BDE-153, BDE-154, BDE-183 and BDE-209, respectively. Mean relative concentrations of p,p'-DDT and o,p'-DDT eliminated via metabolism were less (<50%) than those of p,p'-DDMU, o,p'-DDD, p,p'-DDD, o,p'-DDE and p,p'-DDE (>50%), whereas those of BDE-28, -47

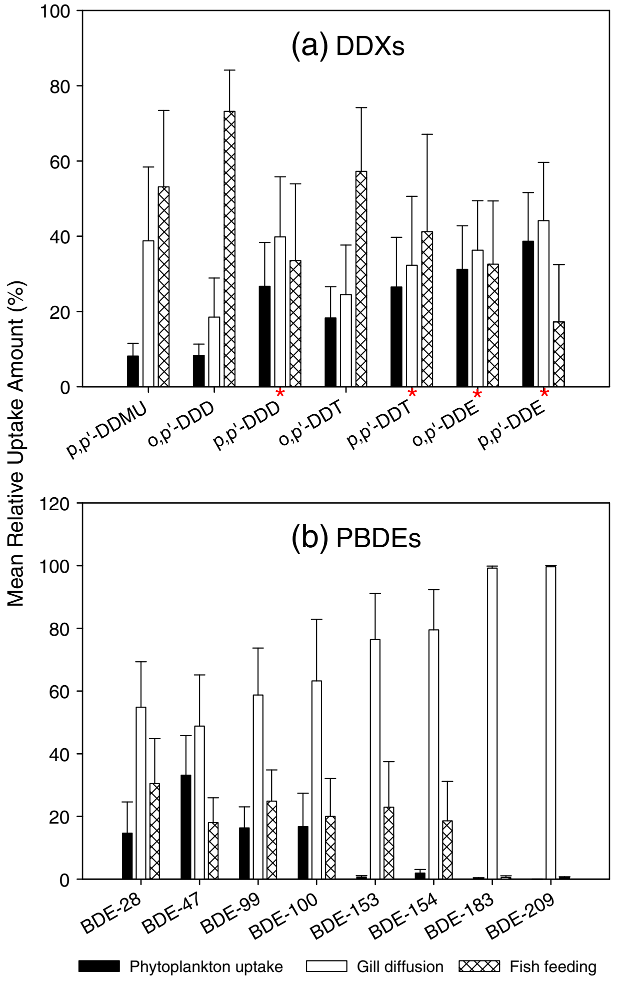

Fig. 2. Mean relative accumulations of PBDE congeners through consumption of phytoplankton, diffusion across gills and through the diet (fish food) for (a) DDXs and (b) PBDEs in farmed freshwater fishes. The bars with symbol * above the *x* coordinate indicate no significant difference ($p > 0.05$) between the three relative uptake amounts for that DDX congener.

and -209 were greater than the other PBDE congeners in farmed marine fishes (Figs. S5 and 6). The relative proportions of DDX and PBDE congeners removed by metabolism from farmed freshwater fishes followed the same pattern as those in farmed marine fishes (Figs. S5 and 6).

The results of simulations, estimated with Equation S52, indicate that at steady state total concentrations of DDXs in farmed freshwater and marine fishes ranged from 1.8 to 17 and 3.3 to 18 ng/g wet wt., respectively. Alternatively, concentrations of PBDEs were predicted to be less than those of DDXs, with estimated ranges of 0.18–4.4 and 0.13–4.3 ng/g wet wt., respectively, in farmed freshwater and marine fishes. These values are less than the measured concentrations. Predicted concentrations of DDXs were from 1.4 to 142 ng/g wet wt. in farmed freshwater fishes and 12–512 ng/g wet wt. in farmed marine fishes. Predicted concentrations of PBDEs ranged from 0.25 to 9.8 ng/g wet wt. in farmed freshwater fishes and 0.16–21 ng/g wet wt. in farmed marine fishes. These results are consistent with steady state being difficult to achieve under environmental conditions. Environmental conditions are more complex than what can be considered in simulation models. Fishes in the environment can be exposed to several contaminants simultaneously. Therefore, it is difficult to maintain constant concentrations of contaminants in fishes. Furthermore, as suggested by Clark et al. (1990), if fish growth occurs, due to growth dilution, true steady state is often not achieved. In fact, the concept of steady state depends on the period being considered. During a short period of time, steady state might be assumed, but during longer periods of time, the assumption of steady state required for the fugacity models cannot be assumed (Mackay, 2001).

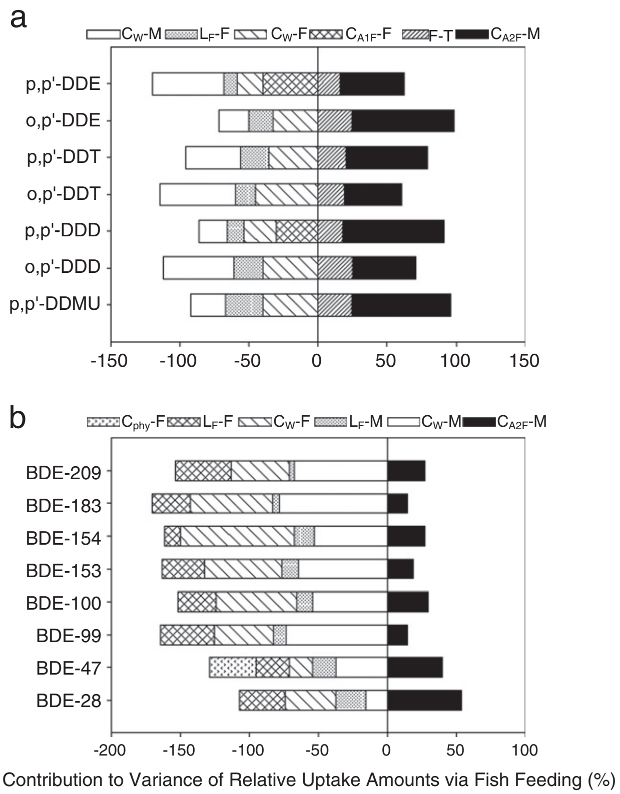

Fig. 3. Sensitivity analysis for main parameters used to estimate relative accumulation amounts via diet of (a) DDX congeners and (b) PBDE congeners based on Equation S47 using Monte Carlo simulation. C_W–M = concentration level in seawater, L_F–F = lipid content of farmed freshwater fish, C_W–F = concentration level in freshwater pond water, C_{A1F}–F = concentration level in phytoplankton from freshwater fish pond, T–F = temperature in freshwater pond water, C_{A2F}–M = concentration level in marine fish feed, C_{phy}–F = Phytoplankton concentration in freshwater fish pond, L_F–M = lipid content of farmed marine fish.

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^{-5}, 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 PHCs 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′*-DDD and 5% 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′*-DDD, 6% for *p,p′*-DDE and 9% 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 assessment results are tentative and qualitative because of insufficient toxicological data for PHCs 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 S7). For carcinogenic hazards, rate of consuming fish in the diet as well as concentrations of *p,p′*-DDD and *p,p′*-DDE in food given to farmed marine fishes and PBDE congeners in seawater are primary parameters (Table S7). Therefore, different risks due to different fish consumption rates are expected for consumers from different regions (Table S4).

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 risk 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.

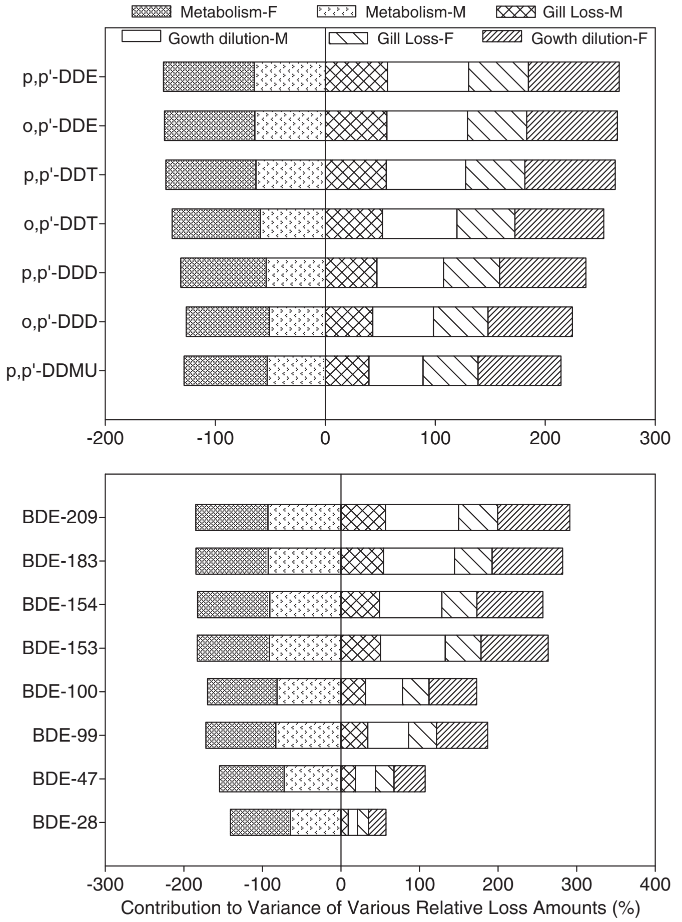

Fig. 4. Sensitivity analysis for value of metabolism half-life time to the variances of various relative losses of DDXs congeners based on Equations S48–S51 using Monte Carlo simulation. Metabolism–F = contribution of metabolism half-life time to the variances of loss via metabolism of farmed freshwater fish. Metabolism–M = contribution of metabolism half-life time to the variances of loss via metabolism of farmed marine fish. Gill loss–M = contribution of metabolism half-life time to variances of loss via gills of farmed marine fish. Growth dilution–M = contribution of metabolism half-life time to the variances of loss via growth dilution of farmed marine fish. Gill loss–F = contribution of metabolism half-life time to the variances of loss via gills of farmed freshwater fish. Growth dilution–F = contribution of metabolism half-life time to the variances of loss via growth dilution of farmed freshwater fish.

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 captured 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 (Guo et al., 2009b), use of compound feed manufactured under controlled conditions should be favored in fish farming.

Table 2
Non-cancer (hazard quotient (HQ) of human exposure to non-carcinogenic exposure limit-reference dose (RfD)) 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^{-5}, respectively.

	Africa	America	Asia	Europe	Oceania
Steady state					
Non-cancer HQs					
o,p′-DDD	0.062	0.084	0.20	0.12	0.053
o,p′-DDE	0.011	0.011	0.030	0.019	0.010
o,p′-DDT	0.033	0.031	0.095	0.054	0.025
p,p′-DDD	0.19	0.23	0.55	0.32	0.13
p,p′-DDE	0.20	0.19	0.61	0.29	0.13
p,p′-DDT	0.060	0.070	0.16	0.098	0.046
BDE-28	0.020	0.019	0.052	0.030	0.011
BDE-47	0.096	0.086	0.24	0.13	0.053
BDE-99	0.032	0.032	0.084	0.044	0.016
BDE-100	0.016	0.015	0.043	0.022	0.0085
BDE-153	0.0024	0.0022	0.0065	0.0033	0.0012
BDE-154	0.0055	0.0049	0.015	0.0078	0.0028
BDE-183	0.0090	0.013	0.028	0.021	0.011
BDE-209	0.0013	0.0013	0.0043	0.0023	0.0012
Cancer hazard level					
DDXs[a]	4.1×10^{-6}	3.7×10^{-6}	7.0×10^{-6}	4.6×10^{-6}	6.4×10^{-6}
BDE-209	3.1×10^{-10}	2.9×10^{-10}	4.6×10^{-10}	4.1×10^{-10}	7.8×10^{-10}
Non-steady state					
Non-cancer HQs					
o,p′-DDD	0.086	0.15	0.34	0.22	0.10
o,p′-DDE	0.037	0.054	0.13	0.078	0.029
o,p′-DDT	0.092	0.16	0.40	0.25	0.11
p,p′-DDD	0.72	**1.3**	**3.2**	**1.8**	0.72
p,p′-DDE	0.50	0.76	**2.0**	**1.1**	0.40
p,p′-DDT	0.52	**1.0**	**2.2**	**1.4**	0.55
BDE-28	0.011	0.0087	0.033	0.013	0.0045
BDE-47	0.092	0.073	0.29	0.11	0.035
BDE-99	0.010	0.011	0.031	0.015	0.0058
BDE-100	0.014	0.013	0.042	0.019	0.0064
BDE-153	0.0032	0.0032	0.0092	0.0045	0.0018
BDE-154	0.012	0.012	0.039	0.018	0.0061
BDE-183	0.025	0.028	0.077	0.051	0.019
BDE-209	0.00016	0.00047	0.00063	0.00091	0.00056
Cancer hazard level					
DDXs[a]	$\mathbf{2.0\times10^{-5}}$	$\mathbf{1.8\times10^{-5}}$	$\mathbf{3.5\times10^{-5}}$	$\mathbf{2.3\times10^{-5}}$	$\mathbf{3.0\times10^{-5}}$
BDE-209	5.7×10^{-11}	1.2×10^{-10}	1.1×10^{-10}	1.7×10^{-10}	3.6×10^{-10}

[a] Sum of *o,p′*- and *p,p′*-DDT, DDD and DDE and *p,p′*-DDMU.

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Appendix A. Supplementary data

Supplementary data to this article can be found online at doi:10.1016/j.envint.2011.04.012.

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