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BIOLOGICAL WASTE AS RESOURCE, WITH A FOCUS ON FOOD WASTE

Source apportionment of DDTs in maricultured fish: a modeling study in South China

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Abstract Fish is one of the most important nutrition sources for humanity. Contaminant exposure risk in fish farming will eventually deliver to the crowd through diet. China is the largest fish producing as well as exporting country, where mariculture plays an important role in fish production, especially in South China. Previous investigations indicated that a variety of compartments in farming areas of South China Sea were polluted by persistent organic pollutants, including DDT (dichlorodiphenyltrichloroethane) and its derivatives, some of which is designated as DDTs. In the present study, Hailing Bay and Daya Bay of Guangdong Province, China, were selected as the study sites and DDTs as the target compounds. A fish enrichment model was developed to assess the relative contributions of various pathways to the mass loadings of DDTs in the fish. Average concentrations (and concentration ranges) of DDTs in various environmental compartments of Hailing Bay and Daya Bay were included in modeling and analysis. Modeling results indicated that fish food and seawater contributed approximately the same proportions for the DDTs in maricultured fish. Antifouling paint was supposed to be the primary source of water DDTs in mariculture zone of Hailing Bay and Daya Bay, which contributed 69 % of the total DDTs to the mariculture water. We suggest that in order to protect people from consuming highly contaminated maricuture zone fish, the most effective and feasible methods are using environment-friendly antifouling paint and applying less polluted fish food in the fish reproduction process.

Keywords Source diagnostics · DDTs · Maricultured fish · Snubnose pompano · Saddletail snapper · Fish enrichment model · South China

Introduction

Globally, more than 31 % of animal protein products are derived from fish (Food and Agriculture Organisation of the United Nations 2014). To tackle the inadequate supply of capture fishery resources, which have been greatly depleted due to overfishing, for meeting the growing market demand, farmed fish have become an important source of fish supply globally (Claret et al. 2012). In 2011, farmed fish contributed to over 40 % of the total worldwide fish production and reached 88.5 % in Asia (Food and Agriculture Organization 2013). China, the world's largest farmed fish producer and exporter, produced approximately 40 million tons of farmed fish in 2011, accounting for more than 50 % of the total global fish production (Chinese Ministry of Agriculture Bureau of Fisheries 2012). In addition, approximately 40 % of the total aquaculture products in China is derived from mariculture (Chinese Ministry of Agriculture 2012), which is mainly located in estuarine areas.

However, due to the extensive use of dichlorodiphenyltrichloroethane (DDT) in China since the 1950s (Xin et al. 2011), both aquaculture zones and ambient aquatic ecosystems have greatly suffered from DDT pollution (Cao et al.

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2007). For example, our previous studies indicated that seawater farmed fish contained higher concentrations of DDTs (designated as the sum of dichlorodiphenyltrichloroethane and its metabolites) than freshwater farmed fish and captured marine fish (Meng et al. 2007a, b, 2009). Further investigations conducted in Hailing Bay and Daya Bay, two net-cage culturing bases in South China, suggested that air, sediment, water, fish food, and biota were polluted with DDTs (Guo et al. 2009a; Yu et al. 2011b, c). Although antifouling paint was identified as a significant source of DDTs in harbors and bays of China (Lin et al. 2009), the contributions of different exposure pathways to the abundant DDTs in maricultured fish have remained to be quantified. It appears that field monitoring alone may not be able to effectively address this issue.

Given the difficulty in diagnosing the source of DDTs in maricutured fish from field surveys, an alternative approach to the issue would be a modeling study with the help of field measurements. In the present study, a receptor model was utilized to examine the source of DDTs in two seawater farmed fish, snubnose pompano (*Trachinotus blochii*) and saddletail snapper (*Lutjanus malabaricus*), which mainly feed on trash fish and compound feed in mariculture zone of South China (Guo et al. 2009a). Hailing Bay and Daya Bay of Guangdong Province, China (Fig. 1) were selected as the study sites, as they have been under intensive investigation for a number of years (Guo et al. 2009b; Liu et al. 2013; Yu et al. 2011a). The results derived from the current modeling efforts were expected to offer in-depth insights into the exposure paths and mechanisms for organic contaminants to enter maricultured fish and ultimately accumulate in humans via fish consumption.

Methodology

Data collection and evaluation

Data related to the occurrences of DDTs in sediment, antifouling paint, atmospheric vapor, atmospheric particulate, water, compound feed, trash fish, algae, snubnose pompano, and saddletail snapper were compiled from the literature (Guo et al. 2009a; Yu et al. 2011b, c), and are presented in the Supplementary Material. All numerical values, except for those directly cited from the literature, were re-computed in the present study. Average concentrations of DDTs obtained from the previous studies (Guo et al. 2009a; Yu et al. 2011b, c) are compiled in Tables S1 and S2. Concentrations of DDTs below reporting limits were set to zero for statistical analysis.

Due to the lack of sufficient DDTs data for all environmental compartments in each mariculture zone, concentrations of DDTs in each matrix from both Hailing Bay and Daya Bay (Guo et al. 2009a, b) were averaged for subsequent analyses. The total number of samples for air, water, surface sediment, snubnose pompano, saddletail snapper, and algae in Haling Bay and Daya Bay was 33, 13, 46, 41, 36, and 8, respectively. In addition, the average concentrations of DDTs in seawater farmed fish food, including 14 trash fish samples and 21 composite feeds of 13 brands, were collected from mariculturing

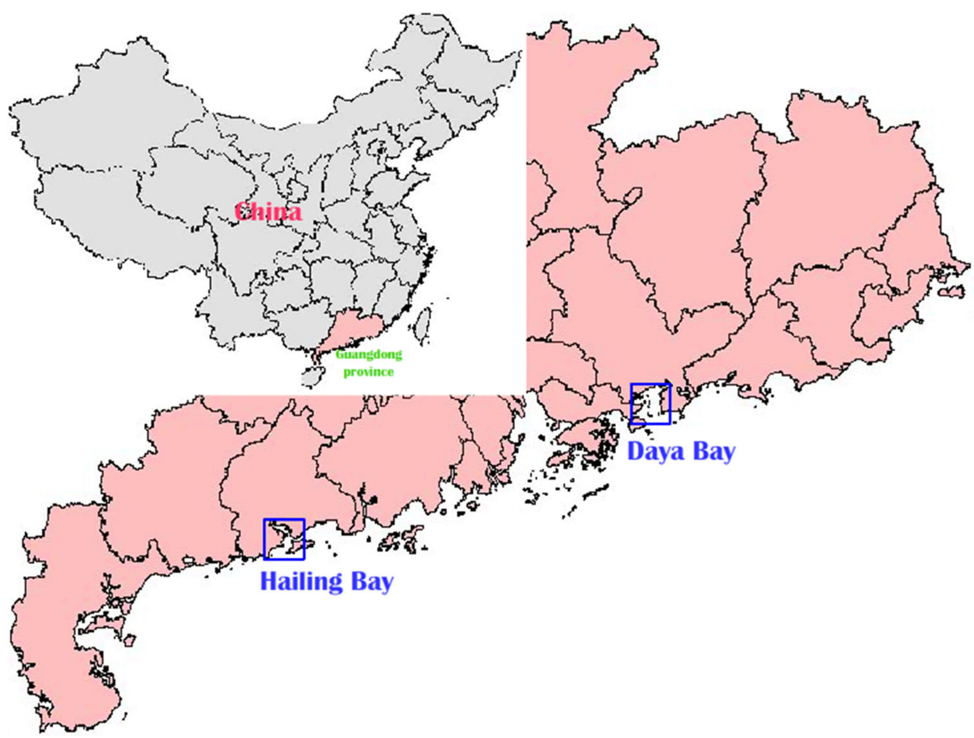

Fig. 1 Map showing the locations of the mariculture zones in Hailing Bay and Daya Bay

zones of Hailing Bay and Daya Bay, South China (Yu et al. 2011a). Data of DDTs are also available for seven antifouling paint products, purchased from a local shop at the coast of Hailing Bay in November 2010 (Yu et al. 2011b). Apart from antifouling paint samples, all other samples were collected between July 2007 and April 2008. Because sufficient number of samples from each type was analyzed in our previous studies, the data thus generated were deemed representative of the average levels of DDTs in different environmental media within the study regions from July 2007 to April 2008.

Model formulation

Receptor models use only the concentrations of chemicals measured at the sources and receptors to quantify source contributions to the receptors (Henry et al. 1984), which may be appropriate for the scenario under investigation. In the present study, the receptors are maricultured snubnose pompano and saddletail snapper in Haling Bay and Daya Bay. A fish enrichment model is developed and used to conduct source diagnostics of DDTs for the two target fish. For a particular compound, the enrichment mechanism in fish can be expressed as (Gobas et al. 1993):

$$\frac{dC_F}{dt} = C_w k_1 + C_d k_d - (k_2 + k_e + k_m + k_g) C_F \quad (1)$$

where C_F and C_w are the analyte concentrations in fish and water, respectively; k_1 is the intake constant through fish gills; k_d is the dietary intake rate constant; and k_2, k_e, k_m, and k_g denote the clearance rates of gill diffusion, feces excretion, metabolism, and growth dilution, respectively. Assuming the analyte concentration in fish is zero at the initial time, C_F can be resolved to give

$$C_F = \frac{\left(1-e^{-k_q t}\right) \times (k_1 C_w + k_d C_w)}{k_q} = \left(1-e^{-k_q t}\right)\left(\frac{k_1}{k_q} C_w + \frac{k_d}{k_q} C_d\right) \quad (2)$$

where C_d is the analyte concentration in fish food and $k_q = k_2 + k_e + k_m + k_g$. Apparently, the relationships among C_F, C_w, and C_d can be multi-linear if time t is assumed as a constant, i.e., within a certain time period, C_F can be further reduced to

$$C_F = \lambda_w C_w + \lambda_d C_d \quad (3)$$

where λ_w and λ_d are the water and feed absorption coefficients, respectively. If there are multiple types of fish food, Eq. (3) can be generalized to

$$C_F = \lambda_w C_w + \lambda_{d1} C_{d1} + \lambda_{d2} C_{d2} + \ldots + \lambda_{dn} C_{dn} \quad (4)$$

where λ_{di} (i=1, 2,…, n) is the weighting coefficient for the ith type of fish food. Equation (4) suggests that the fish enrichment process for the target analyte is characterized by first-order kinetics with negligible biotransformation. In fact, Eq. (4) describes the general form of a first-order chemical mass balance. Based on the chemical mass balance principle, the relative contributions of all possible sources to the loadings of DDTs in fish can be estimated by

$$\begin{cases} \eta_{w-f} = \dfrac{\lambda_w C_w}{\lambda_w C_w + \sum_{i=1}^{n} (\lambda_{di} C_{di})} \times 100\% \\ \eta_{i-f} = \dfrac{\lambda_{di} C_{di}}{\lambda_w C_w + \sum_{i=1}^{n} (\lambda_{di} C_{di})} \times 100\% \end{cases} \quad (5)$$

where $\eta_{w\text{-}f}$ and $\eta_{i\text{-}f}$ denote the relative contributions of water and the ith feed to the analyte loading in fish.

Typically, fish cultured in net cages have no direct contact with sediment, air, and fishing boats, which are defined as indirect compartments for convenience. Analytes released from the indirect compartments primarily enter water first and then can be uptaken by fish, a likely pathway for analytes from indirect compartments to fish. Compartments directly interacting with fish include water and food sources (i.e., composite feed, algae, and trash fish). Transport of analytes between fish and direct compartments can be described by multiple linear relationships in Eq. (4). Similarly, the relationship between water and indirect compartments, which are in direct contact with water but not with fish, can be expressed as:

$$C_w = \lambda_{in,1} C_{in,1} + \lambda_{in,2} C_{in,2} + \ldots + \lambda_{in,m} C_{in,m} \quad (6)$$

where $C_{in,j}$ (i=1, 2,…, m) is the analyte concentration in the jth indirect compartment and $\lambda_{in,j}$ is the weighting coefficient of the jth type indirect compartment, j=1, 2,…., m. The relative contribution of an indirect compartment to the analyte loading in water can be estimated by

$$\eta_{j-w} = \frac{\lambda_{in,j} C_{in,j}}{\sum_{j=1}^{m} \lambda_{in,j} C_{in,j}} \times 100\% \quad (7)$$

where $\eta_{j\text{-}w}$ denotes the relative contribution of the jth indirect compartment to the analyte load in water. Finally, the relative contribution of the jth indirect compartment to the analyte load in fish, $\eta_{j\text{-}f}$, can be characterized by

$$\eta_{j-f} = \eta_{w-f} \times \eta_{j-w} \quad (8)$$

A multiple linear fitting method is adopted to obtain the general weighting coefficients λ_w, λ_{di}, and $\lambda_{in,j}$ in Eqs. (4) and (6) for all DDT analytes using the data in Tables S1 and S2. When the relative contributions in Eqs. (5), (7), and (8) are estimated, analyte concentration is replaced with the sum of DDT concentrations in a particular compartment.

To test the reliability of the fish enrichment model developed, predicted DDT concentrations in water and fish computed by Eqs. (4) and (6) are compared with observed DDT concentrations. The 95 % confidence intervals of predicted values were calculated according to Mackay et al. (2014). On the other hand, the results from source diagnostics of DDTs in snubnose pompano and saddletail snapper are compared with each other. Sensitivity analysis is also conducted with Matlab to identify the most sensitive input parameter concerning the loadings of DDTs in fish.

Results and discussion

Modeling results

Least-square fitting was applied to quantify the regression coefficients in Eq. (4), selecting the concentrations of DDTs in water, composite feed, trash fish, and algae as the independent variables, i.e.,

for snubnose pompano,

$$C_F = 82.2C_w + 0.46C_{feed} + 0.44C_{trash} - 0.11C_{algae} \quad (R^2 = 0.86; p = 0.11) \quad (9)$$

and for saddletail snapper,

$$C_F = 61.4C_w + 0.48C_{feed} + 0.14C_{trash} - 0.08C_{algae} \quad (R^2 = 0.90; p = 0.06) \quad (10)$$

where C_F, C_w, C_{feed}, C_{trash}, and C_{algea} represent the total DDTs concentrations in fish, water, composite feed, trash fish, and algae, respectively. Equations (9) and (10) suggest that water, composite feed, and trash fish contributed positively to the accumulation of DDTs in both fish species, whereas algae had clearance effects. Equations (9) and (10) suggest that transfer of DDTs between two interacting compartments can be characterized with a multiple linear regression method. The concentrations of DDTs in water can be further expressed in terms of antifouling paint, sediment, air vapor, and air particles by

$$C_w = 0.0027C_{paint} + 0.0066C_{sediment} + 0.0018C_{air} - 0.0009C_{particle} \left(R^2 = 0.94; p = 0.003\right) \quad (11)$$

where C_w, C_{paint}, $C_{sediment}$, C_{air}, and $C_{particle}$ represent the total DDTs concentrations in water, antifouling paint, sediment, air vapor, and air particle, respectively.

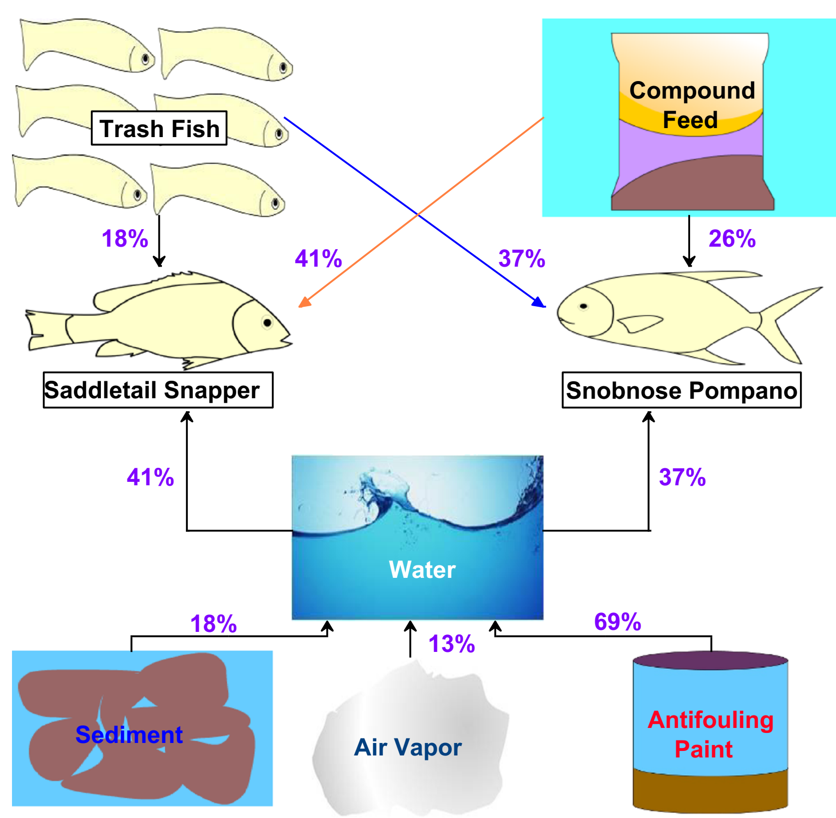

Fig. 2 Modeled intake pathways of DDTs in snubnose pompano and saddletail snapper for Hailing Bay and Daya Bay, South China

Source apportionment of DDTs for direct compartments was calculated with Eq. (5). The results showed that water, composite feed, and trash fish contributed 37 % (2.4–55 %), 26.3 % (6.9–64 %), and 37 % (26–47 %) of the loading of DDTs in snubnose pompano and 41 % (2.9–59 %), 41.4 % (13–78 %), and 18 % (11–25 %) in saddletail snapper (Fig. 2). Relative contributions of fish food to DDTs in snubnose pompano and saddletail snapper were 63 and 59 %, respectively, with a relative difference of 10–20 % compared to water. A previous study found that food and water each contributed 50 % to the body burdens of 1,2,4-trichlorobenzene, 1,2,3,4,5-pentachlorobenzene, and 2,2′,4,4′,6,6′-hexachlorobiphenyl in juvenile rainbow trout (Qiao et al. 2000), similar to our modeling results that water and fish food contributed almost equally to the loadings of DDTs in fish. Apparently, the source profiles are quite different for snubnose pompano and saddletail snapper. Local fishermen indicated that snubnose pomano mainly feeds on composite food, whereas saddletail snapper largely consumes trash fish (Yu et al. 2011b). The inconsistency between the source profiles and feeding inhabits may suggest the occurrence of different biotransformation mechanisms in these two fish species. This corresponds to our previous observations that the compositional profiles were different between saddletail snapper (36 and 29 % of *p,p′*-DDD and *p,p′*-DDE, respectively) and trash fish (21 and 42 % of *p,p′*-DDD and *p,p′*-DDE, respectively) in Hailing Bay (Yu et al. 2011a).

Results from Eq. (7) showed that antifouling paint, sediment, and air contributed 69 % (0.3–96.7 %), 18 % (2.2–47 %), and 13 % (1.8–25 %), respectively, to the total loadings of DDTs in water. The dominance of antifouling paint as the primary source of DDTs in water was consistent with the finding of a previous study (Xin et al. 2011) that DDTs were abundant in wastewater sludge and soil collected from paint factories and shipyards located in Qingdao, Weihai, Shanghai, Guangzhou, and Yangjiang of China. However, the impact of antifouling paint-released DDTs on aquatic systems has not been quantified in previous studies. The present study demonstrated that antifouling paint has contributed predominantly to the occurrence of DDTs in the mariculture zones of Hailing Bay and Daya Bay through seawater.

Application of the fish-enrichment model yielded relative contributions of water-released DDTs at 37 % in snubnose pompano and at 41 % in saddletail snapper. In addition, algae had a clearance effect of DDTs in both fish species. Contributions of aerial deposition to the loadings of DDTs in both snubnose pompano and saddletail snapper were less than 10 %. Antifouling paint indirectly contributed approximately 30 % of DDTs in fish and was apparently the main source of DDTs to water, whereas sediment contributed 6.5 % (0 to 26 %) and 7.3 % (0 to 28 %) to the loadings of DDTs in snubnose pompano and saddletail snapper, respectively, as estimated by Eq. (8). The insignificant contributions of sediment to DDTs in maricultured fish from the two estuarine bays of South China suggest that contaminated sediment is not a major source of DDTs to maricultured fish, although sediment is generally the main destination of pollutants in aquatic environments. This finding is apparently resulted from the currently active release of DDT-containing antifouling paint in the study regions, which may change course once DDTs are not contained in antifouling paint and/or release of antifouling paint is substantially reduced.

Model evaluation

Figures 3 and 4 show that the predicted and measured fish (or water) DDT concentrations were reasonably consistent with each other. Differences between predicted and measured fish (or water) concentrations of DDT components were within an order of magnitude, except for *o,p′*-DDD and *o,p′*-DDE in

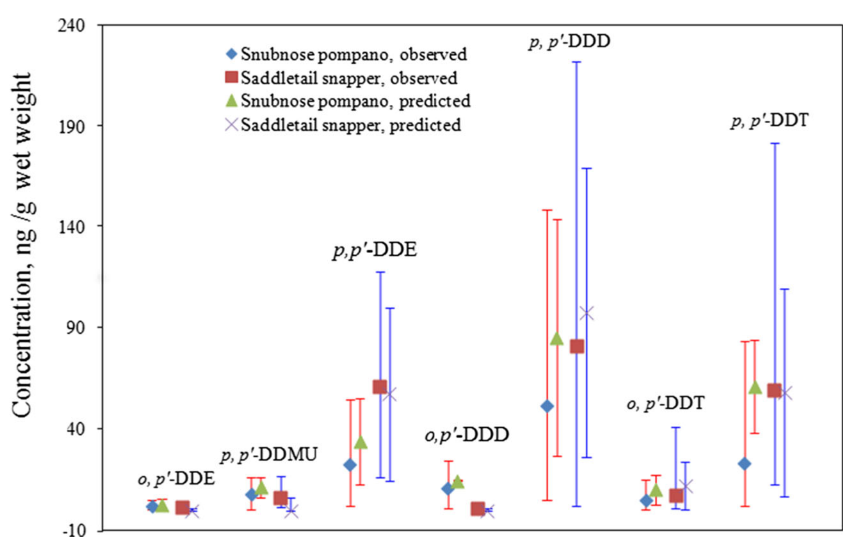

Fig. 3 Comparison between the predicted and observed DDT concentrations with 95 % confidence intervals in snubnose pompano and saddletail snapper from Hailing Bay and Daya Bay; predicted values were obtained with Eqs. (9) and (10)

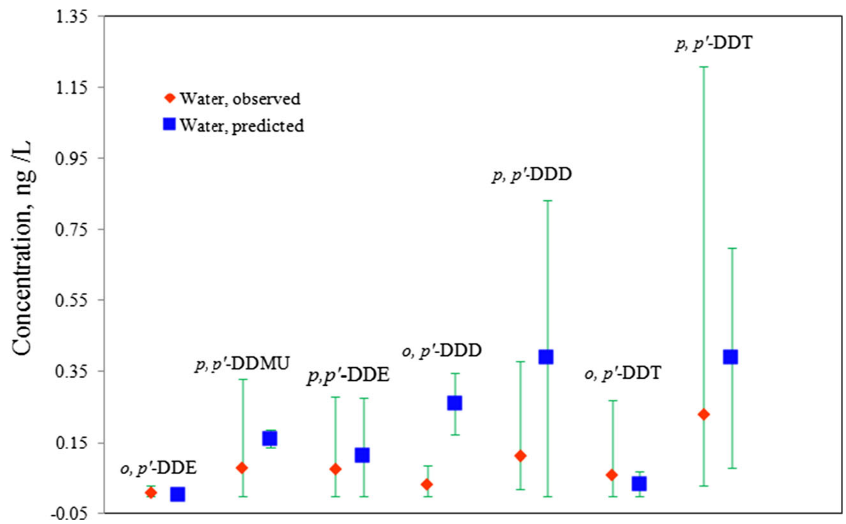

Fig. 4 Comparison between the predicted and observed DDT concentrations with 95 % confidence intervals in water of Hailing Bay and Daya Bay; predicted values were obtained with Eq. (11)

saddletail snapper (Fig. 3), probably due to the large uncertainties of measured values. Therefore, the model established in the present study performed well in describing the accumulation process of DDTs among environment compartments in the mariculture zones of Hailing Bay and Daya Bay.

Sensitivity coefficients of water, sediment, air vapor, compound feed, trash fish, and antifouling paint for snubnose pompano are 0.70, 0.91, 1.3, 0.46, 0.62, and 0.16, respectively. For saddletail snapper, the corresponding values are estimated to be 0.67, 0.88, 1.2, 0.33, 0.81, and 0.15, respectively. Among all parameters, sediment and air vapor posed the strongest influences on the uncertainty of the source diagnostic results. The predicted DDT concentrations in water and the target fish would be considerably improved if more accurate data related to these parameters become available.

It should be noted that some assumptions must be satisfied for the application of the fish enrichment model. First, the compositions of emission sources are maintained constant throughout the sampling duration, which was from July 2007 to April 2008 specific to the present study, and target compounds do not react with each other so that they can be added up linearly. Second, all sources with the potential to contribute to the receptor should be identified and are independent of each other. In the present study, all possible routes for uptake of DDTs in maricutured fish were carefully evaluated and identified. Finally, measurement uncertainties are random, uncorrelated, and normally distributed, which was deemed satisfied in the present study because sufficient number of samples were analyzed in previous studies. Nevertheless, the receptor model developed in the present study is preliminary in nature, and additional field studies are needed to acquire more data so as to further enhance the robustness of model.

Environmental implications

Although the use of DDTs has been officially banned for more than 30 years in China (Lu and Liu 2014), antifouling paint used for fishing boat maintenance still contained abundant DDTs (Yu et al. 2011c; Zheng et al. 2010). Heavy boat traffics in coastal regions may have provoked the release of DDT-containing antifouling paint into the mariculturing zones. Furthermore, it should be pointed out that some fishing nets are probably treated with anti-fouling paint for extension of life span. Released DDTs may eventually enter mariculture fish through bioaccumulation from contaminated sediment and water. In addition, DDTs contained in composite feed and trash fish used as fish foods may also be accumulated in maricultured fish and finally accumulated in humans through fish consumption. Amid the continuous globalization of fish consumption patterns, the results from the present study implicate greatly for human health globally. Although the percentage of mariculture fishery is still slightly lower than that of capture fishery, the amount of aquaculture fisheries in mariculture zones is stupendous (Ababouch 2006). As a result, search for pollution-free fish production modes has become a critical task for meeting the growing global demand for protein sources while minimizing adverse health effects (Icon Group International 2009).

It should be noted that the fish enrichment model developed in the present study is applicable to source diagnostics of organic contaminants not only in marine fish cage culture but also in freshwater fish cage culture. For captured fish, certain information is needed before the fish enrichment model can be used for source diagnostics, i.e., feeding habits, whether the contact with sediment is direct or indirect, and mobility. Nevertheless, the current model is expected to add a new dimension for understanding the potential sources of organic pollutants in captured fish, which is an important factor for protecting human health as capture fishery has remained the global fish supply. Furthermore, our subsequent efforts will focus on developing a generalized version of the fish enrichment model, so as to be able to conduct source diagnostics for other fish species and broaden the knowledge base of organic contamination in mariculture zones.

Conclusions

Fish food and seawater contributed approximately equally to the loadings of DDTs in maricultured fish. Also, antifouling paint was the main source of DDTs in mariculture zone water. The fish enrichment model developed in the present study can be used to assess the impact of sediment-released DDTs on seawater and subsequently on aquaculture. The modeling results suggested that environment-friendly antifouling paint for boat maintenance and pollutant-free fish food should be used in mariculture zones to minimize human health risk due to consumption of maricutured fish.

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