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Modeling the fate of *p,p′*-DDT in water and sediment of two typical estuarine bays in South China: Importance of fishing vessels' inputs☆

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ABSTRACT

Antifouling paint applied to fishing vessels is the primary source of dichloro-diphenyl-trichloroethane (DDT) to the coastal marine environments of China. With the aim to provide science-based support of potential regulations on DDT use in antifouling paint, we utilized a fugacity-based model to evaluate the fate and impact of *p,p′*-DDT, the dominant component of DDT mixture, in Daya Bay and Hailing Bay, two typical estuarine bays in South China. The emissions of *p,p′*-DDT from fishing vessels to the aquatic environments of Hailing Bay and Daya Bay were estimated as 9.3 and 7.7 kg yr^{-1}, respectively. Uncertainty analysis indicated that the temporal variability of *p,p′*-DDT was well described by the model if fishing vessels were considered as the only direct source, i.e., fishing vessels should be the dominant source of *p,p′*-DDT in coastal bay areas of China. Estimated hazard quotients indicated that sediment in Hailing Bay posed high risk to the aquatic system, and it would take at least 21 years to reduce the hazards to a safe level. Moreover, *p,p′*-DDT tends to migrate from water to sediment in the entire Hailing Bay and Daya Bay. On the other hand, our previous research indicated that *p,p′*-DDT was more likely to migrate from sediment to water in the maricultured zones located in shallow waters of these two bays, where fishing vessels frequently remain. These findings suggest that relocating mariculture zones to deeper waters would reduce the likelihood of farmed fish contamination by *p,p′*-DDT.

1. Introduction

Dichloro-diphenyl-trichloroethane (DDT) is an organochlorine pesticide first synthesized in 1874 (Draize et al., 1944). After World War II, DDT was available for commercial sale (WHO, 1979), but then its agricultural use was banned in the 1970s in most countries (Curtis and Lines, 2000). In China, DDT production began in the early 1950s and its use in agriculture was ceased by the Chinese government in 1983 (Qiu et al., 2005). During that period, China produced 0.4 million metric tons of DDT, which accounted for 20% of the global production (Fu et al., 2003; Wong et al., 2005). Because DDT is environmentally persistent, bioaccumulative, and potentially toxic to ecosystems and humans, it was added to the initial list of persistent organic pollutants (POPs) under the Stockholm Convention and its production and usage have been under global restrictions since 2004 (Xu et al., 2013b).

Despite the potential environmental and health risk of DDT, the use of DDT was allowed under circumstances when the benefits of its usage outweigh the associated risk. For example, DDT is widely used in Africa for malaria control as no effective alternatives are available (Ranson et al., 2011). In China, technical DDT was still utilized as an additive to antifouling paints used in fishing ships (Xu et al., 2013c), obviously due to the antifouling effectiveness and affordable prices of technical DDT (Zhang et al., 2014). Antifouling paints used for maintenance of fishing vessels have been identified as the main direct source of DDT detected in Chinese marine areas (MEP, 2013). For example, our previous study (Yu et al., 2011b) found that concentrations of total DDTs (sum of *p,p′*- and *o,p′*-DDT, DDE, and DDD) in some of the commercial antifouling paints purchased from a store in November 17, 2011 along the coast of Hailing Bay of South China were as high as 10 mg g^{-1} wet weight. While all

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DDT and its primary metabolites (*p,p′*- and *o,p′*-DDT, DDE, and DDD) were detected in the samples, *p,p′*-DDT was dominant and accounted for over 70% of the total DDT concentrations (Yu et al., 2011b).

Because fishing activities are frequent in the coastal regions of South China and docks of fishing vessels are often located at the coastal bays, it is important to monitor the impacts of fishing vessels on aquaculture. We are particularly interested in assessing the long-term trends of the state of pollution by DDTs in typical coastal bays of South China, under the scenarios of discontinued and/or continued use of DDT-containing antifouling paints for maintenance of fishing vessels. Our previous studies have accumulated abundant data in two estuarine bays, i.e., Daya Bay and Hailing Bay (Fig. 1), of South China (Fang et al., 2016; Guo et al., 2009; Liu et al., 2013; Yu et al., 2011a,b), which presented the possibility to evaluate the impacts of DDT released from antifouling paints with a comprehensive modeling study. Because of its dominance among all DDT components (including *o,p′*- and *p,p′*-DDT, DDE, and DDD) in antifouling paints, *p,p′*-DDT was selected as the target compound. Using a multimedia environmental fate model under steady state, we quantified the contributions of fishing vessels to the loadings of *p,p′*-DDT in water and sediment of Hailing Bay and Daya Bay and key environmental processes controlling the fate of *p,p′*-DDT in the bays. Using the model under unsteady state, we evaluated the response of the systems to the ongoing elimination of *p,p′*-DDT uses in antifouling paints.

2. Materials and methods

2.1. Environmental and physicochemical properties of p,p′-DDT

Antifouling paint applied to fishing vessels is regarded as the primary source of *p,p′*-DDT in the aquatic environment of the coastal bays. The emission (E; kg yr^{-1}) of *p,p′*-DDT from fishing vessels was estimated by

$$E = \frac{NACF\xi\theta}{\lambda} \cdot 10^{-6} \quad (1)$$

where N is the number of fishing vessels (1140 and 952 for Haling Bay and Daya Bay, respectively); A is the average surface area (m^2) of a typical fishing vessel equivalent to a fin keeled racing craft with the length, width, and height at 6, 3, and 1 m, respectively, and a brushing area of 24 m^2 (Yachpaint, 2015); θ is the ratio of the number of *p,p′*-DDT brushed fishing vessels to the total number of fishing vessels (set as 0.5); C (μg g^{-1}) is the average concentration of *p,p′*-DDT in antifouling paint (5130 ng/g dry weight (dw) (Yu et al., 2011b); F is the annual average brushing frequency and set at 2 based on information from local fishermen; ξ is the leaching rate of *p,p′*-DDT from fishing vessels to seawater and set at 0.7 yr^{-1} (OECD, 2005) for both bays; and λ (m^2 kg^{-1}) is the average stucco area for 1 kg of antifouling paint and set at 5 m^2 kg^{-1} based on information from local fishermen in Hailing Bay.

The physicochemical properties of *p,p′*-DDT used as model inputs are listed in Table S1 of the Supplementary Material ("S" indicates tables and figures in the Supplemental Material afterwards). The areas of Hailing Bay and Daya Bay (Fig. 1) are 180 and 650 km^2, respectively, with active water and sediment depths set at 10 (based on sampling information in Yu et al. (2011a,b)) and 0.01 m (MacLeod et al., 2002), respectively, for both bays. The annual precipitation amounts for Hailing Bay and Daya Bay are 2500 and 1900 mm, respectively. All other non-location specific parameters are taken from the literature (Mackay, 2001; Mackay et al., 2014; Sweetman et al., 2002) and listed in Table S1.

2.2. Environmental fate modeling

The QWASI model first developed by Mackay et al. (1983a,b) was employed in the present study, which deals with chemical mass balance in water and sediment while treating chemical inputs from atmospheric wet and dry depositions as the boundary conditions. Steady-state simulation results were used to represent the contemporary *p,p′*-DDT conditions, whereas unsteady-state results

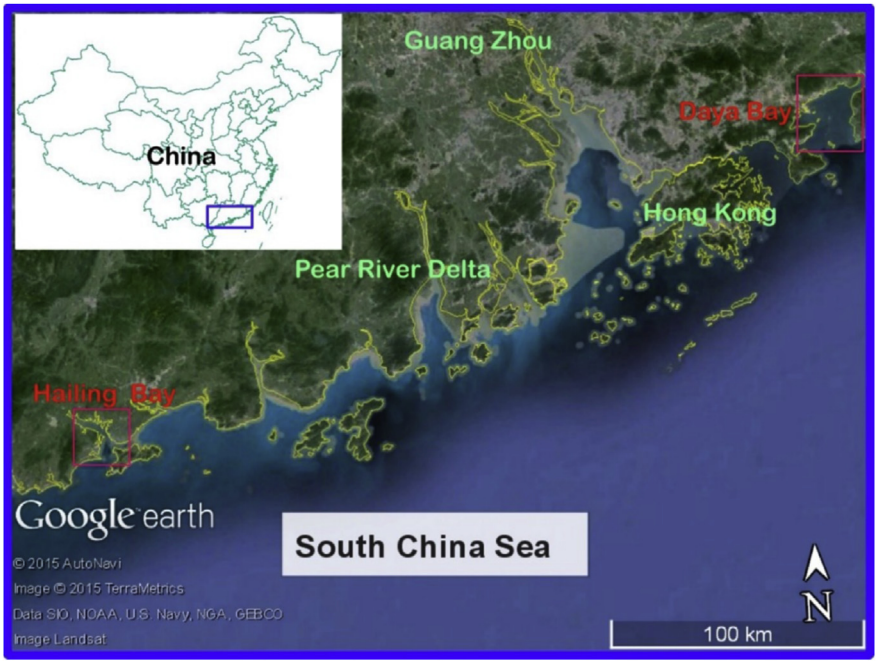

Fig. 1. Locality of Hailing Bay and Daya Bay, two typical coastal bays in South China.

were applied to evaluate the influences of different *p*,*p*′-DDT emission scenarios on the fate and risk of *p*,*p*′-DDT.

Similar to other previous studies using the QWASI model (Mackay and Diamond, 1989; Mackay et al., 2014, 1983a,b; Woodfine et al., 2000; Xu et al., 2013a), the present study also quantified the wet and dry depositions of *p*,*p*′-DDT to water based on measured concentrations of *p*,*p*′-DDT in air. Advective *p*,*p*′-DDT inflow and outflow were not considered because (1) there are no blatant estuaries and no regular flow direction in both bays and (2) there are no scaled estuaries in either bay.

The physical and chemical processes of *p*,*p*′-DDT characterized by the model include burial to deeper sediment, transformation in sediment and water, resuspension to water from sediment, diffusion between sediment and water and between air and water, sediment deposition, volatilization from water to air, and absorption, rain dissolution, and wet and dry particle depositions (Fig. S1). The model adopts a fugacity-based approach and solves mass balance of a chemical in each modeled compartment based on the compartment's fugacity capacity (*Z* values; Table S2) and fugacity-based constants of chemical transfer and transformation rates (*D* values; Table S3) At steady state, the fugacity (*f*; Pa) of *p*,*p*′-DDT in water and sediment can be solved as

$$f_W = \frac{E + f_A(D_V + D_Q + D_C + D_M)}{D_V + D_W + \frac{(D_D+D_T)(D_S+D_B)}{D_R+D_T+D_S+D_B}} \quad (2)$$

and

$$f_S = \frac{f_W(D_D + D_T)}{D_R + D_T + D_S + D_B} \quad (3)$$

where D_V, D_Q, D_C, D_M, D_D, D_T, D_S, D_B, and D_R represent *D* values of volatilization/absorption, dry particle deposition, wet particle deposition, rain dissolution, sediment deposition, sediment to water diffusion/water to sediment diffusion, sediment transformation, sediment burial, and sediment resuspension, respectively. At unsteady-state, the mass balance of *p*,*p*′-DDT in water and sediment can be expressed as

$$\frac{V_W Z_W df_W}{dt} = E + f_A(D_V + D_Q + D_C + D_M) + f_S(D_R + D_T) - f_W(D_V + D_W + D_D + D_T) \quad (4)$$

and

$$\frac{V_S Z_S df_S}{dt} = f_W(D_D + D_T) - f_S(D_R + D_T + D_S + D_B) \quad (5)$$

where V_S and V_W represent the volumes of sediment and water; Z_S and Z_W are the *Z* values of *p*,*p*′-DDT in sediment and water and f_S, f_W, and f_A denote the fugacity values of *p*,*p*′-DDT in sediment, water and air, respectively. Detailed unsteady-state analytical solutions to Equations (4) and (5) can be found elsewhere (Mackay et al., 1983a). The *p*,*p*′-DDT concentration in water (C_W; ng L^{-1}) is calculated as

$$C_w = f_w Z_w M_w \times 10^6 \quad (6)$$

and the *p*,*p*′-DDT concentration in sediment (C_S; ng g^{-1} dw) is calculated as.

$$C_s = f_s Z_s M_w / \rho_s \quad (7)$$

where M_W is the molecular weight of *p*,*p*′-DDT and ρ_S is the sediment density set at 1500 kg m^{-3}.

2.3. Characterization of ecological risk

The environmental risk of *p*,*p*′-DDT was assessed using the Hazard Quotient (HQ) method, based on the model-derived *p*,*p*′-DDT concentrations and screening benchmark values (EPA, 2015), i.e., HQ = EEC/Screening Benchmark. Here, EEC is the estimated *p*,*p*′-DDT concentrations (mean concentrations from Monte Carlo simulation) and Screening Benchmark is generally a non-adverse effect level of *p*,*p*′-DDT. The United States Environmental Protection Agency has set the marine water screening benchmark at 6.5 ng L^{-1} (Water-Benchmark, 2006) and the marine sediment screening benchmark at 1.19 ng g^{-1} (Sediment-Benchmark, 2006) for *p*,*p*′-DDT. The values of HQ at $\leq$ 0.1, 0.1–1, 1–10, and $\geq$10 indicate no hazard, low hazard, moderate hazard, and high hazard, respectively (Lemly, 1996; Yun et al., 2015).

2.4. Sensitivity and uncertainty analyses

Sensitivity analysis was conducted on 20 model input parameters (*X*) to identify the parameters, which the model output (*Y*) is most sensitive to. Sensitivity coefficient (SC) defined as $SC = (\Delta Y/Y)/(\Delta X/X)$ is used to indicate parameter sensitivity (MacLeod et al., 2002; Zhang et al., 2009). Similar to the study of Zhang et al. (2009), modeling was conducted by increasing and decreasing individual parameters by 10%; therefore, $\Delta X/X = 0.2$. Uncertainty analysis was conducted using the Monte Carlo approach (Leterme et al., 2007) to assess possible range of model-derived *p*,*p*′-DDT concentrations in water and sediment of Hailing Bay and Daya Bay. One hundred thousand sets of the input parameters were generated based on the lognormal confidence factors reported in the literature (Mackay et al., 2014; MacLeod et al., 2002; Sweetman et al., 2002) with MATLAB 2014a (MathWorks, Natick, MA, USA) and used for model simulations. The relationships between inputs and outputs, independence, and log-normal distribution of input variables were assumed to be linear, as suggested previously by MacLeod et al. (2002). Results from model simulations were used to generate the distributions of model outputs.

2.5. Analysis of emission control scenarios

To examine how the concentrations of *p*,*p*′-DDT in water and sediment of Hailing Bay and Daya Bay will vary in response to the change in emission rates, the nonsteady-state QWASI model was utilized to simulate the scenario of zero *p*,*p*′-DDT emission. Also, different emission control scenarios were simulated to identify the best strategy for reducing *p*,*p*′-DDT emissions. In one scenario, the temporal trends (50 years) for concentrations of *p*,*p*′-DDT in sediment and water of Hailing Bay and Daya Bay were simulated assuming the emission rate from fishing vessels was zero at a preset year (e.g., the year of 2015). In another scenario, model simulations were performed assuming the current *p*,*p*′-DDT emission rate will remain the same in the future. Other in-between scenarios were also considered (Figs. S2–S5). Measured average *p*,*p*′-DDT concentration in Hailing Bay (or Daya Bay) water (or sediment) was selected as the initial concentration to assess the long-term ecological risk of *p*,*p*′-DDT.

3. Results and discussion

3.1. Model evaluation

Emissions of *p*,*p*′-DDT from fishing vessels in Hailing Bay and Daya Bay estimated with Equation (1) were 9.3 and 7.7 kg yr^{-1}, respectively, which were the basic data used in subsequent modeling efforts. When normalized to the bay areas, the emissions

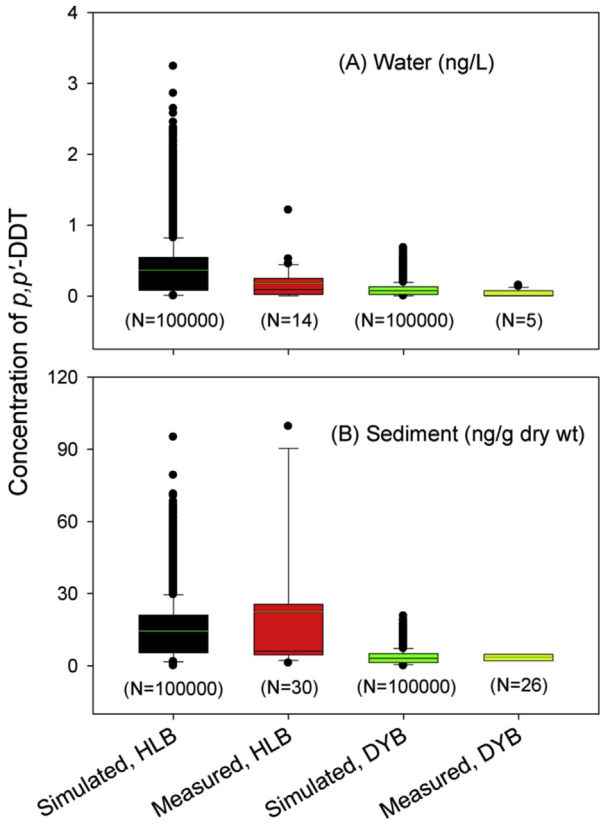

Fig. 2. Comparison of simulated and measured *p,p′*-DDT concentrations in (A) water and (B) sediment of Hailing Bay (HLB) and Daya Bay (DYB).

of *p,p′*-DDT in Hailing Bay and Daya Bay became 5.17×10^{-4} and 1.18×10^{-4} kg ha^{-1} yr^{-1}, respectively, which were significantly low than the emission of *p,p′*-DDT (4.65 kg ha^{-1} yr^{-1}) estimated by Wong et al. (2005) in China in 1985 when DDT was still used as a pesticide. However, both study regions, particularly Hailing Bay, can be characterized as a current "hot spot" of *p,p′*-DDT due to abundant *p,p′*-DDT contents in water and sediment (Yu et al., 2011a,b) and should be dealt with swiftly and adequately. Actions for controlling *p,p′*-DDT pollution in coastal areas include, for instance, the development and popularization of environmentally friendly, affordable, and effective antifouling paints.

With the estimated emissions of *p,p′*-DDT mentioned above, the model can be evaluated by a comparison of modeled and field-measured data. Modeled steady-state concentrations of *p,p′*-DDT in water and sediment of Hailing Bay and Daya Bay (Fig. 2) were comparable to measured concentrations obtained between June and December of 2007 (Yu et al., 2011a,b). Simulated and measured mean *p,p′*-DDT concentrations were 0.36 and 0.18 ng L^{-1} in Haling Bay water, 14.5 and 23.9 ng g^{-1} dry weight in Hailing Bay sediment, 0.087 and 0.038 ng L^{-1} in Daya Bay water, and 3.5 and 3.4 ng g^{-1} dry weight in Daya Bay sediment; the differences between the simulated and measured data were within a factor of 3 (±200%). Moreover, field-observed intervals were braced by simulated intervals, i.e., simulated and observed intervals of *p,p′*-DDT concentrations were 0–3.8 and 0.005–1.2 ng L^{-1} in Hailing Bay water, 0.003–102 and 1.1–99.5 ng g^{-1} dry weight in Hailing Bay sediment, 0–0.93 and 0.005–0.15 ng L^{-1} in Daya Bay water, and 0.001–24.5 and 0.87–5.3 ng g^{-1} dry weight in Hailing Bay sediment. To conclude, the simulated results were in good agreement with field measurements results in both Hailing Bay and Daya Bay. Thus, the environment fate of *p,p′*-DDT in Hailing Bay and Daya Bay was appropriately described by the localized steady-state QWASI model.

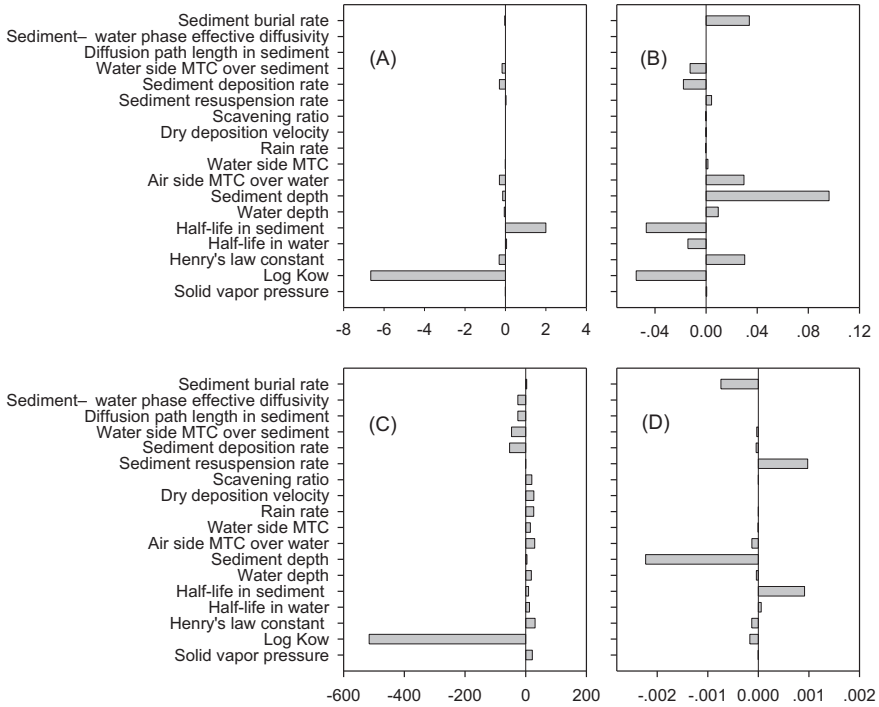

Fig. 3. Sensitivity of modeled *p,p′*-DDT concentrations in (A) water of Hailing Bay (B) sediment of Daya Bay (C) water of Daya Bay and (D) sediment of Daya Bay to model parameters. The sensitivity is quantified using sensitivity coefficient defined as $SC = (\Delta Y/Y)/(\Delta X/X)$.

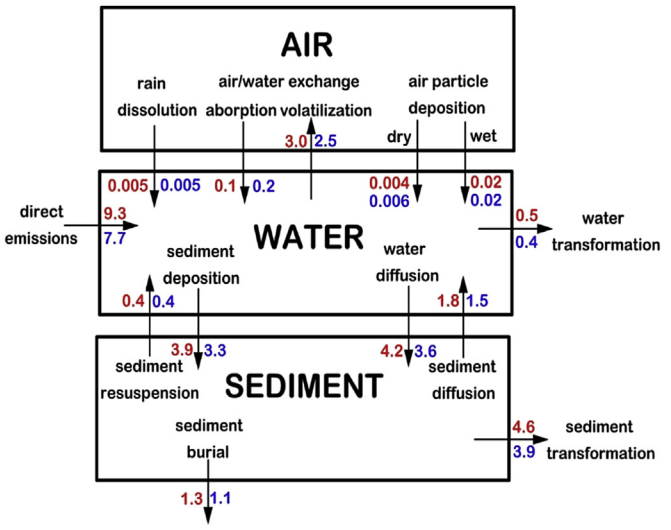

Fig. 4. Environmental fate of *p,p′*-DDT in Haling Bay (data in red color) and Daya Bay (data in blue color) estimated based on steady-state model simulations. Unit: kg yr^{-1} (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.).

3.2. Implications of sensitivity and uncertainty analyses

Results of sensitivity analyses intended to examine the influence of individual parameters on *p,p′*-DDT concentrations in water and sediment are shown in Fig. 3. In Hailing Bay water, the octanol–-water partition coefficient (log K_{ow}) was the most sensitive parameter, followed by the half-life of *p,p′*-DDT degradation in sediment, Henry's Law constant, sediment deposition rate, air-side mass transfer coefficient (MTC) over water, and water-side MTC over sediment. Parameters exerting significant influences on *p,p′*-DDT concentrations in sediment were sediment depth, log K_{ow}, half-life of *p,p′*-DDT degradation in sediment, sediment burial rate, Henry's Law constant, air side MTC over water, and sediment deposition rate. In Daya Bay, log K_{ow} was the most sensitive parameter dictating the concentration of *p,p′*-DDT in water, while the most sensitive parameter in controlling the concentration of *p,p′*-DDT in sediment was the sediment depth. Uncertainty analyses indicated that the intervals of observed *p,p′*-DDT concentrations in both water and sediment of these two bays were encompassed by the simulated intervals (Fig. 2).

3.3. Environmental fate of p,p′-DDT

Fig. 4 illustrates the environmental fate of *p,p′*-DDT in Hailing Bay and Daya Bay. Under the steady-state conditions, nearly 99% of *p,p′*-DDT in the aquatic environment was distributed in sediment of both Hailing Bay and Daya Bay, consistent with the strong hydrophobicity of *p,p′*-DDT (log $K_{ow} = 6.91$). Also, the fugacity of *p,p′*-DDT was higher in water than in sediment in both bays, indicating a flux direction from water to sediment. Sediment transformation was the most important process for removal of *p,p′*-DDT from the aquatic system. Finally, the residence times of *p,p′*-DDT were 15 days and 52 years in water and sediment, respectively, at both bays. The procedures for estimating the residence time of chemicals in water and sediment can be found in page 208 of Mackay (2001).

Two key processes governing the mass balance of *p,p′*-DDT in the water of Hailing Bay were particle deposition (3.9 kg yr^{-1}) and water-sediment diffusive exchange (4.2 kg yr^{-1}). These two processes accounted for 34% and 36% of *p,p′*-DDT removal from the water compartment. In sediment, chemical transformation (4.6 kg yr^{-1}) and diffusion from sediment to water (1.8 kg yr^{-1}) were two key processes, accounting for 57% and 22% of *p,p′*-DDT emitted. The overall residence time of *p,p′*-DDT within the entire Hailing Bay system was estimated as 45 years, with chemical transformation in sediment and evaporation from water to air contributing to 49% and 32% of the total amount of *p,p′*-DDT removed from the system.

Similarly, sediment deposition and chemical diffusion from water to sediment were also the two key processes of *p,p′*-DDT in Daya Bay water. Via these two processes, 34% and 37% of *p,p′*-DDT emitted to water were removed. In Daya Bay, sediment deposition and chemical diffusion from water to sediment were two key processes of *p,p′*-DDT, removing 57% and 22% of *p,p′*-DDT emitted to water. For the entire Daya Bay system, the overall residence time (46 years) was close to that in Hailing Bay (45 years). In addition, chemical transformation in sediment and evaporation from water were identified as the two most important processes for removing *p,p′*-DDT in this area, with removal efficiencies of 49% and 32%, respectively.

The steady-state modeling results showed that in both Hailing Bay and Daya Bay, *p,p′*-DDT tended to transport from water to sediment. By comparison, our previous modeling study in Hailing Bay and Daya Bay found that DDTs tended to transport from sediment to water in the mariculture zone (Fang et al., 2016). Contaminated sediment contributed 6.5% (0–26%) and 7.3% (0–28%) to the loadings of DDTs in snubnose pompano (*Trachinotus*

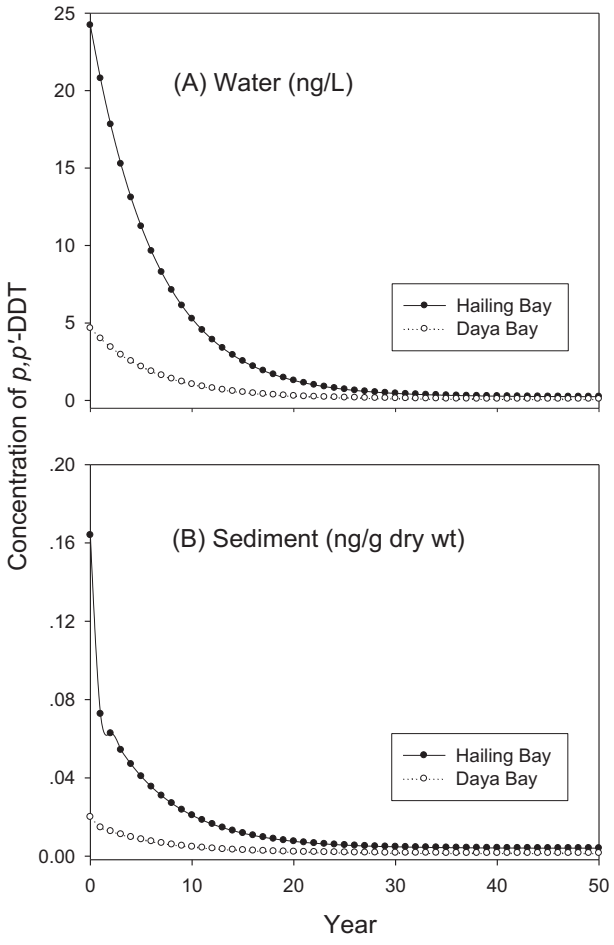

Fig. 5. Simulated long-term trends of *p,p′*-DDT concentrations in (A) water and (B) sediment of Haling Bay and Daya Bay after the use of *p,p′*-DDT in antifouling paint for maintaining fishing vessels is banned.

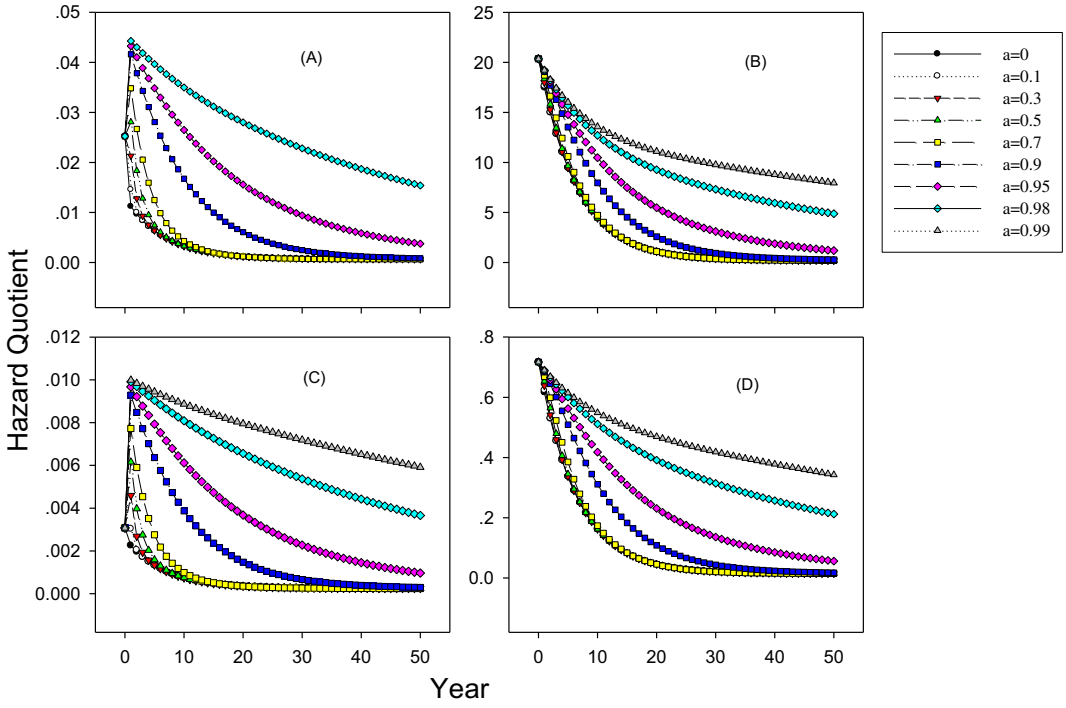

Fig. 6. Simulated long-term trends for ecological risk (in terms of hazard quotients) of *p,p′*-DDT in water and sediment of Haling Bay and Daya Bay under different source control scenarios. The *p,p′*-DDT emissions over time are assumed to follow $E(t) = E(0)\,a^t$, where a = 0, 0.1, 0.3, 0.5, 0.7, 0.9, 0.95, and 0.98 and are plotted here. The reduction year of *p,p′*-DDT in fishing vessels is set to zero. $E(0) = 9.3$ kg yr^{-1} for Hailing Bay and $E(0) = 7.3$ kg yr^{-1} for Daya Bay. (A) Hailing Bay water; (B) Hailing Bay sediment; (C) Daya Bay water; and (D) Daya Bay sediment.

blochii) and saddletail snapper (*Lutjanus malabaricus*), respectively (Fang et al., 2016). On the other hand, antifouling paint contributed 69% (0.3–96.7%) of the total loadings in Hailing Bay and Daya Bay water (Fang et al., 2016). Our field visits noticed that the mariculture zones in Hailing Bay and Daya Bay are mainly located in shallow waters, where fishing vessels frequently remain. These findings indicated that contaminant loadings in maricultured fish may be reduced if mariculture zones are reallocated in deeper waters, resulting in reduced human exposure to *p,p′*-DDT through fish consumption.

3.4. Variability of *p,p′*-DDT concentrations and risk under various source control scenarios

Unsteady-state model simulations were conducted to investigate how concentrations and environmental risk of *p,p′*-DDT in sediment and water will change when the use of *p,p′*-DDT in antifouling paint is regulated. Here we focused on an optimistic scenario under which the source of *p,p′*-DDT is completely eliminated and its emission becomes zero (Fig. 5). Results for other *p,p′*-DDT regulation scenarios are presented in Figs. S2–S5.

When emissions of *p,p′*-DDT become zero in Hailing Bay, it would take 1 and 5 years for the concentrations of *p,p′*-DDT in water and sediment to decline to 50% of its initial values. It would take 4 and 5 years to accomplish the same goal in Daya Bay. Concentrations of *p,p′*-DDT in water will decrease by 87%, 95%, and 97% in Hailing Bay and 76%, 88%, and 91% in Daya Bay after 10, 20, and 30 years. In addition, concentrations of *p,p′*-DDT in sediment will decrease by 78%, 95%, and 98% in Hailing Bay and 78%, 94%, and 97% in Daya Bay after 10, 20, and 30 years. It would take about 30 years for *p,p′*-DDT in the aquatic system to reach a new steady-state in both Haling Bay and Daya Bay. Besides, it would take 4 to more than 100 years for *p,p′*-DDT concentrations in water and sediment of these two bays to decline by half of the initial values for all other scenarios (Figs. S2–S5).

Hazard quotients for *p,p′*-DDT in Hailing Bay and Daya Bay under different *p,p′*-DDT source control scenarios are shown in Fig. 6. At present, HQs for *p,p′*-DDT in water (sediment) of Haling Bay and Daya Bay were 0.025 (20.4) and 0.003 (0.72), respectively. Due to its high hydrophobicity, no risk of *p,p′*-DDT in water of either Hailing Bay or Daya Bay was observed. Hazard quotients of *p,p′*-DDT in sediment were higher than those in water, likely because *p,p′*-DDT tends to accumulate in the organic matter fraction of sediment. In Hailing Bay, *p,p′*-DDT in sediment posed high environmental risk. Scenario analysis suggested that it would take 5–29 years to reduce the hazard level of *p,p′*-DDT in sediment of Hailing Bay to a safe value, depending on how efficient and soon control actions are taken to eliminate DDTs in antifouling paint.

4. Conclusions

The present study demonstrated a convenient method with the use of existing information to estimate the emissions of *p,p′*-DDT in Hailing Bay and Daya Bay. The modeling efforts based on a QWASI model suggested that fishing vessels were the dominant source of *p,p′*-DDT in Hailing Bay and Daya Bay, with emission rates of *p,p′*-DDT from fishing vessels being estimated as 9.3 and 7.7 kg yr^{-1}, respectively. Besides better regulation of the use of *p,p′*-DDT in antifouling paints, relocation of mariculture zones to deeper waters of the bays would substantially lower human exposure to *p,p′*-DDT through dietary intake of maricultured fish. Finally, the current method can potentially be applied for estimating the emission rates of pollutants from fishing vessels globally.

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Appendix A. Supplementary data

Supplementary data related to this article can be found at http://dx.doi.org/10.1016/j.envpol.2016.02.052.

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