Environ Sci Pollut Res (2013) 20:8425–8434
DOI 10.1007/s11356-013-1809-4

Biota–sediment accumulation factor (BSAF), bioaccumulation factor (BAF), and contaminant levels in prey fish to indicate the extent of PAHs and OCPs contamination in eggs of waterbirds

C. K. Kwok · Y. Liang · S. Y. Leung · H. Wang · Y. H. Dong · L. Young · J. P. Giesy · M. H. Wong

Received: 8 February 2013 / Accepted: 6 May 2013 / Published online: 24 May 2013
© Springer-Verlag Berlin Heidelberg 2013

Abstract Samples of pond sediment, fish, and shrimp were collected from the Ramsar site at Mai Po marshes, Hong Kong (south China), and samples of pond sediment, fish, and shrimp, as well as eggs of water birds (Chinese Pond Herons (*Ardeola bacchus*) and Little Egrets (*Egretta garzetta*)), were collected from two smaller wetland sites at Jiangsu Province (mid-China), between 2004 and 2007. Accumulation levels of polycyclic aromatic hydrocarbons (PAHs) and organochlorine pesticides (OCPs) in the biota were used to calculate biota–sediment accumulation factor (BSAF) and bioaccumulation factor (BAF). For fish and shrimp, BSAFs of OCPs (3.8–56) were greater than those of PAHs (0.12–6.3). BSAFs and BAFs of 11–79 and 4–34, respectively, were registered for OCPs in eggs of the birds and were greater than those for PAHs (0.11–1.5 and 0.02–1.3, respectively). Assuming that fish were the main prey of the birds, greater bioaccumulation of OCPs was detected for both bird species (BAFs=4.5–34), while accumulation of PAHs was only detected in Little Egret (BAF=1.3). A significant linear relationship ($p<0.01$) was observed between concentrations of OCPs in bird eggs and in the prey fish. The present study provides a new possibility of using OCP levels detected in prey fish to predict the extent of OCPs contamination in eggs of waterbirds including the endangered species, as a noninvasive method.

Responsible editor: Philippe Garrigues

Electronic supplementary material The online version of this article (doi:10.1007/s11356-013-1809-4) contains supplementary material, which is available to authorized users.

C. K. Kwok · Y. Liang (✉) · S. Y. Leung · M. H. Wong (✉)
Croucher Institute for Environmental Sciences, and Department of Biology, Hong Kong Baptist University, Hong Kong SAR, People's Republic of China
e-mail: yliang@hkbu.edu.hk
e-mail: mhwong@hkbu.edu.hk

H. Wang · Y. H. Dong
Institute of Soil Science, Chinese Academy of Sciences (ISSAS), Nanjing, Jiangsu, People's Republic of China

L. Young
World Wide Fund Hong Kong, Mai Po Nature Reserve, Hong Kong SAR, People's Republic of China

J. P. Giesy
State Key Laboratory on Marine Pollution Control, City University of Hong Kong and Xiamen University, Hong Kong, People's Republic of China

J. P. Giesy
Biomedical Sciences and Toxicology Center, University of Saskatchewan, Saskatoon, SK, Canada

J. P. Giesy
Zoology Department and Center for Integrative Toxicology, Michigan State University, East Lansing, MI 48824, USA

J. P. Giesy
Environmental Science Program, Nanjing University, Nanjing, People's Republic of China

Keywords Bioaccumulation factor · Biota–sediment accumulation factor · Persistent organic pollutants · Wetland · Ardeids

Introduction

In Hong Kong, deterioration of the Inner Deep Bay Mai Po Ramsar site due to the presence of heavy metals and persistent organic pollutants (POPs) has received much attention and concern during the past decade. Water birds, being the top predators in the wetland ecosystem (Furness 1993), are major inhabitants of this wetland, and Ardeids are the major resident species and are therefore readily exposed to the bioaccumulatable pollutants. It is commonly known that dichlorodiphenyldichloroethylene (DDE), a metabolite of dichlorodiphenyltrichloroethane (DDT), caused eggshell thinning and the subsequent severe population declines in a number of bird species in America and Europe (Stokstad 2007; Vos et al. 2000). In our region, it has been reported that contamination of organochlorine pesticides (OCPs) in eggs of Ardeid species may adversely affect breeding and fledging success of the species (Connell et al. 2003; Lam et al., 2008).

It has been revealed that 4- to 6-ring aromatic compounds are the most toxic to birds (including embryos, young birds, and adult birds). For adult and young birds, the adverse effects include reduced egg production and hatching. However, there is limited information on the increasing concentrations of environmental polycyclic aromatic hydrocarbons (PAHs) on bird populations (Albers 2006). Eggs of golden eagles from the Scottish borders were analyzed for 52 PAHs, and most of them were detected in eggs and were at likely embryotoxic concentrations (Pereiara et al. 2009). Analysis of total of 77 eggs of Kentish Plover (*Charadrius alexandrinus*) from ten breeding sites of the Iberian Atlantic coast after a major oil spill that happened in November 2002 revealed that in general, concentrations of PAHs decreased from 2004–2006, but the pattern of PAH accumulation in 2007 was mainly caused by the tetra and pentacyclic compounds from forest fires that occurred during the summer of 2006 (Vidal et al. 2011).

Although collection of eggs for contaminant analysis is regarded as a comparatively noninvasive method, operational difficulties in sample collection, such as the strict regulations for collecting live samples (especially endangered species) imposed by local authorities (in particular Hong Kong, where the Ramsar site is managed by World Wide Fund for Nature), may hinder investigations of the extent of contamination threatening the health and survival of water birds. Therefore, if concentrations of contaminants of concern in other ecological compartments, especially those in the prey food of water birds (e.g., fish and shrimp) and sediments, can serve as indicators of contamination in bird eggs, the degree of invasiveness and difficulty in sampling could be reduced.

It is hypothesized that biota–sediment accumulation factor (BSAF) and bioaccumulation factor (BAF) values of sediments and prey fish in wetlands, where water birds seek shelter and food, could be used to predict concentrations of OCPs and PAHs in bird eggs of Ardeids species, via analyzing BSAF or BAF values from appropriate correlations/regressions obtained. The major objectives of the present study were to (1) monitor the concentrations of OCPs and PAHs in fish species and pond sediments collected from the Ramsar site at Mai Po marshes, Hong Kong, south China, and based on the data obtained, calculate BSAF values for OCPs and PAHs in different fish species, (2) monitor the concentrations of OCPs and PAHs in fish species and eggs of two species of Ardeids from another wetland reserve from Jiangsu Province, mid-China, and based on the data obtained, calculate both BSAF and BAF values for OCPs and PAHs in bird eggs, and to (3) quantify the relationship between concentrations of OCPs in bird eggs, with those in prey fish, based on the data obtained from the present study, as well as those from other relevant studies.

Materials and methods

Site descriptions

Biota and sediment samples were collected from two different sites: the Ramsar site at Mai Po marshes, Hong Kong, south China and two small isolated wetlands, namely Gu Cheng (GC) county and Shang Hu (SH) county of Jiangsu province (JS), mid-China (Fig. 1). Mai Po marshes (N22'29.920, E114'02.682), together with the Inner Deep Bay area located at the northeast of Hong Kong, is the largest piece of wetland habitats, including an intertidal mudflat, a mangrove swamp and some traditionally operated shrimp ponds (named as gei wais locally, with gates facing the seaward side to trap aquatic organisms to be grown inside the ponds), where samples were collected for this study. The area has attracted a large number of migratory birds every winter from the far north, to seek shelters and food. This is due to the traditional practice of draining pond water, resulted in a lower water level and sometimes exposed pond mud, which attracted birds to feed on fish and benthic organisms. The birds regularly visiting the site included a few endangered species, notably Black-faced Spoonbill (*Platalea minor*). The Ramsar site at Mai Po is administrated by the World Wide Fund for Nature, Hong Kong, and the area is relatively far away from urban centers. However, there is a danger that Ramsar site is gradually degraded because of urbanization and environmental pollutants from the Pearl River Delta, receiving domestic and industrial effluent discharged from major cities along the

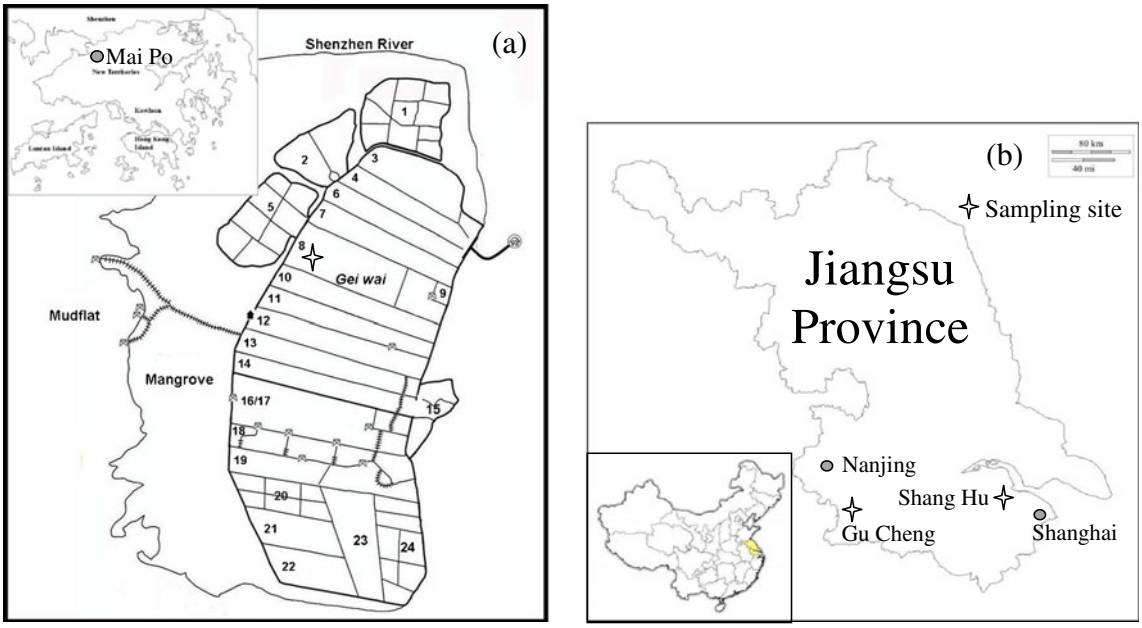

Fig. 1 Maps of sampling location in **a** Mai Po Ramsar site, Hong Kong; and **b** Gu Cheng and Shang Hu county, Jiangsu province

river, entering the Ramsar site from the north and northwest (Liang and Wong 2003; Agriculture, Fisheries and Conservation Department 2011).

The two small isolated wetlands are located at GC county (31′14.639′N, 119′00.184′E) and SH county (31′39.118′N, 120′41.483′E) of JS province, mid-China. Jiangsu Province is a flat and low-laying plain, with a well-developed irrigation system for its agriculture, with rice, wheat, maize, sorghum as the major crops and cash crops including bamboo, tea, medicinal herbs, and gingko. The province is also a center for producing silk and freshwater fish. It is also one of the most densely populated regions in China and has been a hot spot for economic development, with 21 "economic and technological development zones", with major products such as electronic equipment, chemicals, and textiles (http://www.uvista.com/en/jiangsu/suzhou.htm). Therefore, the birds visiting the sites may be threatened by the higher levels of OCPs and PAHs due to the rapid regional development.

Sample collection and pretreatment

In Hong Kong, biota samples including gray mullet (*Mugil cephalus*), tilapia (*Oreochromis mossambicus*), snakehead (*Channa maculata*), crucian carp (*Carassius carassius*), mud carp (*Cirrhinus molitorella*), tenpounder (*Elops saurus*), Indo-Pacific tarpon (*Megalops cyprinoids*), gei wai shrimps (*Metapenaeus* sp.), and together with the corresponding surface sediment samples (0–5 cm) (with $n=3$) were collected in both gei wais and fish ponds at Mai Po (a Ramsar site), Hong Kong, during Aug–Sept 2005 and 2006. Pond draining is usually operated in late autumn to winter (Young and Melville 1993) so the current sampling should represent the equilibrium state of the pond system. In addition, crucian carp, shrimp, eggs of Chinese Pond Herons (CPH, *Ardeola bacchus*), and Little Egrets (LE, *Egretta garzetta*), and surface sediment (0–5 cm) of fish ponds (with $n=3$) were collected from two different sites, namely GC county and SH county of JS province, China in June 2005 and May 2007. Sediment and biota samples of each site were collected within an area of 5×5 m to reduce variations. The fish samples and shrimp samples were divided into different sizes. There were three size classes for fish: small (S): 40–80 g, medium (M): 81–330 g, and large (L): 331–770 g. Shrimps were divided into four size classes: S: <6 g, M: 6–10 g, L: 1–15 g, and extra large (XL): >15 g.

All sediment and biota samples were kept at 4 °C in airtight plastic bags immediately after collection and during transport to the laboratory. Sediments were weighed to determine their mass (wet weight) using a top-loading balance, then frozen at −20 °C overnight before freeze-drying for approximately 1 week. Freeze-dried sediment was weighed again (dry weight; dw), sieved (2 mm), and then kept for later determination of PAHs and OCPs. All the sieved dry samples were stored in a desiccator.

Lengths and weights of fish and shrimp samples were recorded using a ruler and a top-loading balance, respectively. Muscles of fish and shrimp were dissected, their mass determined, and then frozen and freeze-dried. Freeze-dried muscle of the same fish/shrimp species collected from the same site was homogenized using a homogenizer (Kinematica, Polymix A10, Switzerland). The dry samples were placed into a desiccator for storage. Whole eggs of LE and CPH (from China) were weighed, and the egg content was poured into a beaker for freeze-drying.

Soxhlet extraction and cleanup

Freeze-dried sediment (4 g)/biota (0.5–1 g) samples were weighed into cellulose thimbles (ADVANTEC®, Grade 84) and soxhlet extracted using 80 mL of 1:1 (v/v) pesticide grade acetone and dichloromethane (DCM) (Labscan Asia Co., Ltd.) at 65 °C for 18 h, according to the standard method 3540C (United State Environmental Protection Agency USEPA 1996a). Approximately 0.2 g of certified reference material 105–100 (PAHs and pesticide-contaminated soil, certified by USEPA) (Resource Technology Corporation, USA) was also employed for quality assurance during soxhlet extraction. After evaporating the solvent mixture to approximately 5 mL using a rotatory evaporator (BÜCHI, Rotavapor® R-114), solvent extracts of the sediment samples were added with approximately 0.1 g of activated copper powder (Riedel-de Haën, prewashed with 1 mol/L hydrochloric acid and washed with double-distilled water, acetone, and DCM) for sulfur removal. Determination of lipid contents of biota was based on the method described by Antoniadou et al. (2007). Briefly, solvent extract was made up to 5 mL, in which 1 mL of the extract (in glass vials) was weighed and then evaporated (for the organic solvent) at 70 °C overnight. The weight difference prior to and after evaporation was used for lipid content determination. The florisil cleanup method (standard method 3620B, United State Environmental Protection Agency USEPA 1996b) was used to remove impurities in the extracts. Florisil columns were activated at 150 °C for 4 h before use. Depending on the cleanup efficiency, five to six 15 mL portions of 99 % n-hexane (Labscan Asia Co., Ltd.) were used to elute the extracts in the cleanup process. The cleaned extracts were concentrated to less than 2 mL using a rotatory evaporator. The final extracts were added with 10 μL internal standard (320 ng/g acenaphthene, chrysene, and phenanthrene) and then made up to 2 mL using n-hexane, which were then stored in 2-mL vials at 4 °C before measurement.

Gas chromatography and mass spectrometry

Sixteen PAHs and 15 OCPs, identified as priority pollutants by the USEPA due to their toxic, mutagenic, and carcinogenic characteristics (United State Environmental Protection Agency USEPA 1996c), in the soxhlet extracts of both sediment and biota samples were analyzed. The 16 PAHs included naphthalene, acenaphthylene, acenaphthene, fluorene, phenanthrene, anthracene, fluoranthene, pyrene, benz(a)anthracene, chrysene, benzo(b)fluoranthene, benzo(k)fluoranthene, benzo(a)pyrene, indeno(1,2,3-cd)pyrene, dibenzo(a,h)anthracene, and benzo(g,h,i)perylene. The 15 OCPs included hexachlorobenzene, heptachlor, aldrin, trans-chlordane, cis-chlordane, trans-nonachlor, cis-nonachlor, dieldrin, endrin, *p*,*p*'-DDE, *o*,*p*'DDE, *o*,*p*'-DDD, *p*,*p*'-DDD+*o*,*p*'DDT, *p*,*p*'-DDT, and mirex. The standard method 8270C (semivolatile organic compounds by gas chromatography/mass spectrometry) (United State Environmental Protection Agency USEPA 1996c) was used to quantify the organic pollutants. A Hewlett Packard 6890 GC system together with a mass selective detector, connected with a 30 m×0.25 mm×0.25 μm DB-5 capillary column (J & W Scientific Co. Ltd., USA) were used for analyses. The carrying gas for chromatography was helium. A reference material of soil CRM105 was used, and the recoveries of different analytes in CRM105 were: naphthalene (112 %), acenaphthene (93.3 %), fluorene (96 %), phenanthrene (86.2 %), anthracene (102 %), fluoranthene (74 %), pyrene (93.2 %), benzo(b+k)fluoranthene (111 %), dieldrin (58.3 %), *p*,*p*'-DDE (99.3 %), and *p*,*p*'-DDD (118 %). The detection limits for all species of PAHs were rounded up to 20 ng/g, while those of OC pesticides were 1–10 ng/g (1 ng/g: *p*, *p*'-DDE; 2 ng/g: heptachlor, aldrin, dieldrin, endrin, and *p*,*p*'-DDD; 10 ng/g: *p*,*p*'-DDT).

Calculation of BSAFs and BAFs and statistical analysis

BSAFs and BAFs are calculated as below:

$$\mathrm{BSAF} = C_{\mathrm{b1}}/C_s \quad (1)$$

$$\mathrm{BAF} = C_{\mathrm{b1}}/C_{\mathrm{b2}} \quad (2)$$

Where: C_{b1}, C_{b2} are the concentrations of a given contaminant in biota samples, while C_{s} is the concentration of the contaminant in sediment. SPSS version 10 was employed for statistical analyses. One-way ANOVA and independent sample *t* test were used to compare any differences ($p<0.05$) in PAH and OCP concentrations in the samples. Regression analysis was performed to determine any linear relationship ($p<0.05$) between BSAFs/BAFs for PAHs and OCPs in biota and the octanol–water partition coefficients (K_{ow}) of the corresponding organic compounds, as well as the relationship between OCPs in eggs of Ardeids and prey fish. Values of log K_{ow} for individual PAH and OCP are listed as below: naphthalene (3.37), acenaphthene (3.92), fluorene (4.18), phenanthrene (4.57), anthracene (4.54), fluoranthene (5.22), pyrene (5.18), benz(a)anthracene (5.91), chrysene (5.6) (Mackay et al. 1997), acenaphthylene (3.5) (Department of Environmental Quality and Louisiana 2003), trans-chlordane (6.22), cis-chlordane (6.1), trans-nonachlor (6.35), cis-nonachlor (6.08) (Simpson et al. 1995), and *p*,*p*'-DDE (6.76) (United State Environmental Protection Agency USEPA 2000). The total organic carbon content (TOC) in aquaculture ponds is known to be generally stable (Boyd et al., 1994), therefore, 2 % TOC was used for calculation of BSAFs as reported in an earlier study concerning polychlorinated biphenyls (PCBs) contamination in Mai Po (Liang et al. 1999).

Results and discussion

Concentrations of OCPs and PAHs in fish and shrimp collected from Mai Po

Tables S1 and S2 (supplementary materials) show that the concentrations (lipid base) of OCPs and PAHs in muscles of various fish and shrimp collected from the shrimp ponds (gei wais) of Mai Po, Hong Kong varied among species. In general, the concentrations in fish were significantly ($p<0.05$) higher than those in shrimp, while there were no significant differences ($p>0.05$) among the same fish species of different sizes (for all gray mullet, tilapia, and mud carp).

Similar finding was also observed between gray mullets and gei wai shrimps, in which ∑OCPs in the former were about 2.6 times greater than the later (Wong et al. 2006). Such difference can be explained by the higher position of fish in food chains of the pond ecosystem. It is generally known that POPs can be accumulated in aquatic organisms over a long period of time (United Nations Environment Program 2005), with longer half-lives, e.g., half-life of DDT may reach 10–15 years (Mörner et al. 2002). Fish would, because of their longer life span, i.e., 11 years for tilapia (Masterson 2007), accumulate greater concentrations of ∑OCPs and ∑PAHs than did shrimp (<16 months for gei wai shrimp, Leung 1997). Although the present results did not indicate size difference in terms of both contaminants detected in fish, a previous study showed that the size of tilapia affected PAH bioaccumulations in the viscera of small (506 ng ∑PAH/g), < medium (591 ng), and < large (854 ng) individuals (Liang et al. 2007). Such different findings highlight the variability in the pattern of PAH bioaccumulation between different body compartments of fish.

According to the present results, feeding habit and habitat may affect the extent of bioaccumulation. In general, higher OCPs and PAHs were noted in Indo-Pacific tarpon (carnivore: eating small fish, shrimps, and insects), tilapia (omnivore: a wide variety of food items including plant-based materials, algae, insect larvae, etc.), and mud carp (omnivore: mainly detritus, water plants, insects, and benthic organisms), while lower concentrations in gray mullet (herbivore: mainly plant materials and phytoplankton), tenpounder (carnivore: small fish and shrimps), and crucian carp (carnivore: mainly insects and zooplankton) (FishBase 2013). It seemed apparent that carnivores and omnivores tend to accumulate these pollutants more efficiently than herbivores and detritus feeders (with the sole exception of tenpounder). This can probably explain the greatest concentrations of ∑OCPs (13,000 ng/g, lipid weight (lw)) and ∑PAHs (27,000 ng/g, lw) detected in muscle of Indo-Pacific tarpon, regarded as an intermediate carnivore, which consumes mainly (about 45 %) fish and crustaceans in its diet (Ley 2007). Our previous study also indicated that black bass (*Micropterus salmoides*), a strict carnivore commonly reared in the Pearl River Delta region, usually accumulates greater concentration of organochlorines (Zhou et al. 1999).

Fortunately, the present results revealed lower concentrations of ∑DDTs and ∑PAHs in fishes of Mai Po than those from other urban centers in the Pearl River Delta. ∑DDTs in tilapia and crucian carp of Mai Po were approximately 16 to 37 % of those from Guangzhou and Shipai, while ∑PAHs in tilapia of Mai Po were about 71–78 % of those from these two cities (Kong et al. 2005). However, it was observed that *p*,*p*'-DDE seemed to be the most dominated congener of the OCPs detected in most of the fish species, which was possibly due to the large amount of DDTs produced and used in China in the past (Wong et al. 2005).

Values of BSAFs for ∑OCP and ∑PAH in fish and shrimp of Mai Po

The BSAFs for ∑OCPs and ∑PAHs varied among fish and shrimp (Table 1), and the values for ∑OCPs were greater than those for ∑PAHs. The greatest ($p<0.05$) were found in tilapia, while the least ($p<0.05$) in gray mullets.

Table 1 Biota–sediment accumulation factors for ∑PAHs and ∑OCPs in different fish and shrimp collected in Mai Po Ramsar site

	∑OCPs Mean±SD	∑PAHs Mean±SD
Omnivores		
Tilapia (l)	33±27 ^{bc}	3±2.5 ^{abc}
Tilapia (s)	56±18 ^{d}	6.3±1.4 ^{d}
Mud carp (l)	46±22 ^{cd}	3±1.5 ^{abc}
Mud carp (m)	13±6.7 ^{ab}	1.7±1.0 ^{abc}
Carnivores		
Crucian carp (s)	6.6±1.3 ^{a}	0.23±0.03 ^{a}
Tenpounder (l)	5.3±1.0 ^{a}	0.12±0.01 ^{a}
Snakehead (l)	9.8±2.4 ^{a}	0.91±0.1 ^{ab}
Herbivores and plankton feeders		
Gray mullet (s)	4±0.5 ^{a}	0.13±0.02 ^{a}
Gray mullet (l)	3.8±0.8 ^{a}	0.12±0.03 ^{a}
Shrimp (s)	7.2±3.5 ^{a}	4.1±2.7 ^{cd}
Shrimp (m)	5.9±3.6 ^{a}	3.9±3.2 ^{bcd}
Shrimp (l)	8.2±5.1 ^{a}	2.5±1.6 ^{abc}
Shrimp (XL)	5.3±1.1 ^{a}	2.7±1.1 ^{abc}
Feral eel[1]	1–70	
Fish[2]	0.7–8.6	
Sunfish[3] ($\times 10^{-3}$)		0.01–5
Lake trout[4] ($\times 10^{-3}$)		0.1–7

For BSAFs in a column, means followed by the same letter are not significantly different at 0.05 probability level according to Duncan test. Refer to (Table S1, supplementary materials) for the sizes of fish and shrimp. [1] Van der Oost et al. (1996), [2] Wong et al. (2001), [3] Thomann and Komlos (1999), [4] Burkhard and Lukasewycz (2000)

BSAFs values of OCPs were in the range of 3.8 to 56 (Table 1). The present BSAFs for ∑OCPs are greater than the national-scale value reported for fish in USA (0.7–8.6 for eight different OCPs, Wong et al. 2001) but within the range reported for feral eel (*Anguilla anguilla*) (approximately 1–70, Van der Oost et al. 1996) (Table 1). In addition, significantly greater ($p<0.05$) BSAFs for ∑OCPs were observed in bottom-dwelling species, such as tilapia (33–56) and mud carp (13–46). Feeding habits of these omnivorous fish, e.g., consuming sediments as one of their food items, may contribute to the greater BSAFs (Zhou et al. 1998; Zhou and Wong 2000). Organisms that are in close vicinity to bottom sediment, such as tilapia and mud carp, may therefore accumulate greater concentrations of OCPs as reflected by the greater BSAFs. Such greater BSAFs observed for tilapia, for instance, are also reported in the accumulation of DDT (24.1, which is about three times of the value of catfish; Leung et al. 2010) and heavy metals (0.19–7.48, which are about 40–85 % greater than the values of catfish; Adeniyi et al. 2008). In contrast, species that do not usually consume sediments as their major diets, such as shrimps and gray mullets, had lesser BSAF values (3.8–8.2). Shrimps, for example, were reported to feed mainly on algae and zooplankton, but not sediment (Zhou and Wong 2000). These results showed that the sediment-feeding behavior would largely affect the extent of OCP bioaccumulation in aquatic organisms.

Alternatively, BSAFs for ∑PAHs in fish and shrimp of Mai Po (0.12–6.3) are notably higher than the values reported for sunfish (*Lepomis* sp.) (1×10^{-5}–5×10^{-3}, Thomann and Komlos 1999) and lake trout (*Salvelinus namaycush siscowet*) (1×10^{-4}–7×10^{-3}, Burkhard and Lukasewycz 2000) (Table 1), and would probably indicate some extent of bioaccumulation in the local species. In contrast to ∑OCP, shrimps seem to accumulate PAHs more readily than the fish in Mai Po (except tilapia and mud carp). Similar findings of varied BSAFs among different species of fish and shrimp were reported by Maruya and Lee (1998) that bioaccumulation of ∑PCBs was observed in striped mullets (*M. cephalus*, BSAF=3.1), while metabolism/elimination was observed in grass shrimps (*Palaemonetes pugio*, BSAF=0.28) and sea trout (*Cynoscion nebulosus*, BSAF=0.81). This can be explained by the sediment-ingesting behavior of striped mullets during feeding, which may lead to the much greater BSAF in the study. The

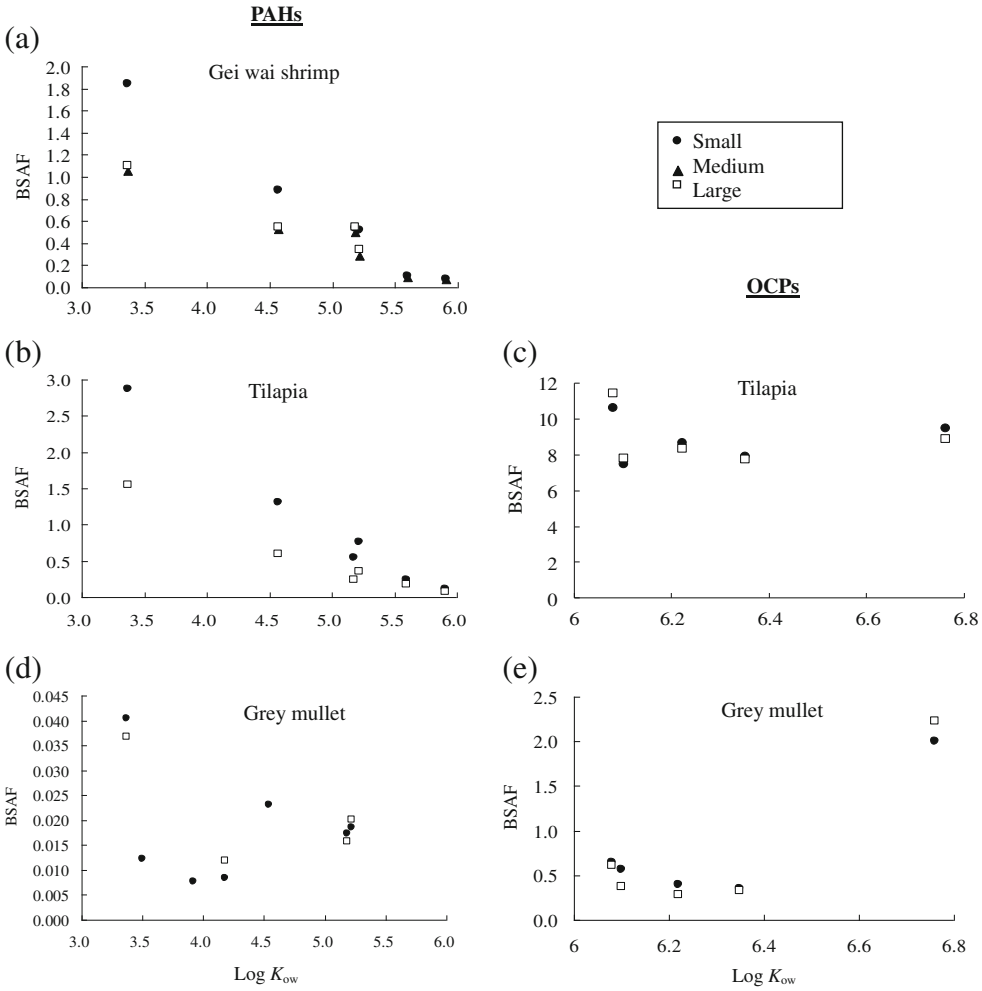

Fig. 2 Plots of biota–sediment accumulation factors (BSAFs) for PAHs and OCPs against log K_{ow} for gei wai shrimps (**a**), tilapia (**b**, **c**), and gray mullet (**d**, **e**)

present results are consistent with the observation that BSAFs values for ∑PAHs in sediment-ingesting species (tilapia and mud carp, 6.3 and 3.0, respectively) were considerably greater than those of other fish species, particularly predatory fish such as snakehead (0.91), a strict carnivore feeding on other fish to acquire a diet that contains relatively great proportions of protein (Paripatananont 2002). This feeding habit allows less exposure to sediment for snakeheads compared to tilapia and mud carp, thus leading to a lesser BSAF.

Significant linear relationships were found between log K_{ow} of individual PAH and OCP and the corresponding BSAFs for fish and shrimp (Fig. 2). The BSAFs for individual OCP in gray mullets were positively correlated with log K_{ow} (BSAF= 2.336 log K_{ow} −13.94, r^2=0.731, p=0.002) (Fig. 2e), while no linear relationship was found for tilapia (Fig. 2c). In contrast, negative linear relationships were found between values of log K_{ow} of individual PAH and the corresponding BSAFs in large (BSAF=−0.589 log K_{ow}+3.43, r^2=0.956, p=0.001) and small (BSAF=−1.12 log K_{ow}+6.54, r^2=0.980, p<0.001) tilapia (Fig. 2b) but not in gray mullets (Fig. 2d). Also, strong negative correlations were found between BSAFs for individual PAH and corresponding log K_{ow} in shrimps (BSAF=−0.501 log K_{ow}+3.01, r^2=0.825, p<0.001) (Fig. 2a). As for tilapia, different regressions for shrimp of different sizes were also obtained (small: BSAF=−0.716 log K_{ow}+4.22, r^2=0.988, p<0.001; medium: BSAF=−0.392 log K_{ow}+2.37, r^2=0.945, p=0.001). The result is not unexpected, as more lipophilic compounds (e.g., high-ringed PAHs with high K_{ow} values) tend to bind more tightly with the sediments containing higher organic matter, resulting in less partition from sediments to water, and therefore lower bioavailability to and bioaccumulation in fish. However, the low-ringed PAHs with low K_{ow} values appeared to be more mobile between sediments and water, contributing to higher bioaccumulation in fish.

Table 2 BSAFs and BAFs for ∑PAHs and ∑OCPs in eggs of Ardeids collected from Jiangsu province, China

		∑PAHs	∑OCPs
BSAF			
Location	Biota		
SH	Egg of LE	1.5±0.5	79±44
GC	Egg of CPH	0.11±0.01	40±9.3
SH	Egg of CPH	0.41±0.18	11±1.2
BAF			
Location	Biota		
SH	Egg $_{LE}$-c. carp	1.31	25
SH	Egg $_{LE}$-shrimp	0.03	0.94
GC	Egg $_{CPH}$-c. carp	1.0	34
SH	Egg $_{CPH}$-c. carp	0.92	4.5
SH	Egg $_{CPH}$-shrimp	0.02	0.17

Locations: Shang Hu (*SH*) county, Gu Cheng (*GC*) county, both at Jiangsu province. Birds: Little Egret (*LE*), Chinese Pond Heron (*CPH*), *c. carp*: crucian carp

Values of BSAF are shown in mean±SD, while those for BAF are mean values. Lipid contents of crucian carp, eggs of LE, and CPH were adopted from Antoniadou et al. (2007) and Lam et al. (2007) (5.1, 5.7 and 5.1 %, respectively) for calculations of BSAFs and BAFs

Birds and polycyclic aromatic hydrocarbons

Values of BSAFs and BAFs for PAHs and OCPs in eggs of Ardeids from Jiangsu province

Bioaccumulation of OCPs and PAHs into eggs of Little Egret and Chinese Pond Heron of Jiangsu province, China varied greatly (Table 2), while BSAFs for ∑PAHs were much less than those for ∑OCPs. Species difference was observed, as BSAFs for eggs of LE were approximately 2–14-fold greater than those of Chinese Pond Heron. In particular, OCPs and PAHs in Ardeid eggs had BAF values greater than one, assuming that fish were a major prey of the birds. The crucian carp was chosen as a representative species, as it was reported as a major prey of the Ardeids species in China (Dong et al. 2002; Ruan et al. 2003), and it constituted up to 30 % of the Ardeids diets in Korea (Sang et al. 2001). In contrast, lesser values of BAFs for OCPs and PAHs were found, when shrimp was assumed to be the main prey of the waterbirds.

Extrapolation of pollutants in eggs of Ardeids from prey fish

The present study showed a general pattern of bioaccumulations of ∑OCPs and ∑PAHs in eggs of Little Egret and Chinese Pond

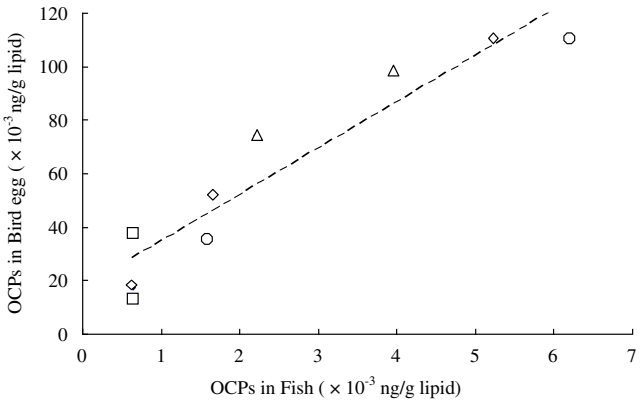

Fig. 3 OCP concentrations in eggs of Ardeids against prey fish obtained in Jiangsu province and other wetlands. Jiangsu province (this study); Tai Lake, China (Dong et al. 2004); Mai Po (Connell et al. 2003); Pakistan (Sanpera et al. 2003). Ardeids species include Little Egret, Chinese Pond Heron and Black-crowned Night Heron (*Nycticorax nycticonax*). OCP concentrations of eggs and fish from other studies were converted to lipid weight, in which lipid contents of 6.5, 5.6, and 5.1 % were used for eggs of LE, Black-crowned Night Herons (Lam et al. 2007), and fish (Antoniadou et al. 2007), respectively. Data of Crucian carp in the present study (1,800 ng/g, lw) was integrated with the data of Connell et al. (2003), in which OCPs were only analyzed for eggs of LE and Black-crowned Night Herons

Heron of Jiangsu province. However, ∑OCPs seemed to be more readily bioaccumulated in eggs of these water birds than ∑PAHs, as the BAFs for OCPs were greater than those for PAHs (Table 2). Moreover, Little Egret seemed to be more susceptible to accumulate OCPs than Chinese Pond Heron did, as the accumulation ratios for the former were generally higher than those for the latter. A similar pattern was also observed for PAHs (Table 2). When comparing the results of the current study with those of other studies, lower concentrations of ∑OCPs were observed in eggs of Little Egret (2,300 ng/g, dw) and Chinese Pond Heron (3,200 ng/g, dw) collected from Tai Lake, China (Dong et al. 2004), as well as in eggs of Little Egret collected from two Ramsar sites in Pakistan (1,100 and 3,400 ng/g, dw in Haleji Lake and Taunsa Barrage, respectively) (Sanpera et al. 2003).

Since Ardeids mostly feed on fish, it is expected that the relatively great concentrations of OCPs in eggs were resulted from intake of contaminated fish. A significant linear relationship between concentrations of OCPs in Ardeids eggs and prey fish (Fig. 3) was obtained via integration of the results from this study and the studies of Connell et al. (2003), Sanpera et al. (2003), and Dong et al. (2004) ($OCPs_{Ardeids\ eggs}=17.38\ OCPs_{prey\ fish}+16{,}822$, $r=0.944$, $p<0.0001$). These studies reported bioaccumulation of OCPs in both Ardeid eggs and prey fish. Integration of data from these studies allow analysis of the relationship between the contaminant levels in both biotic compartments and enhance the feasibility to construct a regression model as shown in Fig. 3. This finding allows estimation of OCP contaminations in eggs of Ardeids by simply analyzing OCPs in prey fish, which avoids disturbance to the wildlife, by minimizing the demand of egg sample collection. This is particularly useful for evaluating the contamination status of endangered species, in which collection of live samples is impossible.

By substituting concentration of ∑OCPs in crucian carp (1,800 ng/g, lw (Table S1) into the regression model, a greater concentration of total OCPs (48,000 ng/g, lw) in eggs of Ardeids of Mai Po was obtained. The concentration of total OCPs in crucian carp and other fish species in the present study (Table S1) derived using this method fell into the range reported by Connell et al. (2003) for Little Egrets (14,000–58,000 ng/g lw, original data in wet weight were converted to lipid weight using lipid content of 6.5 % [Lam et al. 2007]) of Mai Po. This finding indicated that there is no large discrepancy between the calculated and measured OCP concentrations in eggs of Ardeids of Mai Po.

The BAFs of PAHs were approximately the same for the $Egg_{CPH-carp}$ relationship observed in the two locations of Jiangsu province (Gu Cheng County (1.0) and Shang Hu County (0.92)) (Table 2). This indicates that the pattern of PAH bioaccumulation in the same predator–prey relationship could be similar among similar habitats and may allow the use of BAFs/BSAFs in Jiangsu province to extrapolate the degree of PAH contamination in birds of Mai Po.

Conclusion

The present results revealed that gray mullets, tenpounder, and Indo-Pacific tarpons contained greater concentrations of ∑PAHs and ∑OCPs than other fish and shrimp species of Mai Po. Greater BSAFs for ∑PAHs and ∑OCPs were found for bottom-dwelling and sediment-feeding species, including tilapia and mud carp, indicating ∑OCPs and ∑PAHs could be readily bioaccumulated in the bodies of these fish species. The relatively great BAF and BSAF values for ∑OCPs and ∑PAHs in eggs of Ardeids of Jiangsu province indicated that the pollutants can be bioaccumulated in birds via exposure to both contaminated sediment and fish (BAFs and BSAFs >1). A significant linear regression was obtained between ∑OCP concentrations in Ardeids eggs and prey fish, and the model appeared to be useful for estimating ∑OCP contamination in eggs of Ardeids of Mai Po, which could also potentially be employed in extrapolating OCP contamination in eggs of endangered water bird species. Nonetheless, for a more comprehensive investigation in the future and application of the results, more sampling sites and samples (especially bird egg samples) are essential to verify the models derived from this study.

Acknowledgments The authors thank Mr. Tse Man Fung and Mr. Bena Smith for his assistance for field sampling. Financial support from Science Faculty Strategic Research of Hong Kong Baptist University (no. FRG/03-04/II-51) and Environment and Conservation Fund of Hong Kong Environmental Protection Department (ECF project no. 16/2003) is gratefully acknowledged. Prof. J.P. Giesy was supported, in part, by an at large chair professorship at the Department of Biology and Chemistry and Research Center for Coastal Pollution and Conservation, City University of Hong Kong.

References

Adeniyi AA, Yusuf KA, Okedeyi OO (2008) Assessment of the exposure of two fish species to metals pollution in the Ogun river catchments, Ketu, Lagos, Nigeria. Environ Monit Assess 137:451–458

Agriculture, Fisheries and Conservation Department (2011) Mai Po Inner Deep Bay Ramsar Site Management Plan. AFCD, Hong Kong, China

Albers PH (2006) Birds and polycyclic aromatic hydrocarbons. Avian Poult Biol Rev 17:125–140

Antoniadou V, Konstantinou IK, Goutner V, Sakellarides TM, Albanis TA, Bintoudi E (2007) PCB levels and accumulation pattern in waterbird eggs and in their prey at Lake Kerkini, a northeastern Mediterranean wetland of importance. Arch Environ Contam Toxicol 53:249–260

Boyd CE, Tanner ME, Madkour M, Masuda K (1994) Chemical characteristics of bottom soils from freshwater and brackish-water aquaculture ponds. J World Aquacult Soc 25:517–534

Burkhard LP, Lukasewycz MT (2000) Some bioaccumulation factors and biota–sediment accumulation factor in polycyclic aromatic hydrocarbons in lake trout. Environ Toxicol Chem 19:1427–1429

Connell DW, Fung CN, Minh TB, Tanabe S, Lam PKS, Wong BSF, Lam BHW, Wong LC, Wu RSS, Richardson BJ (2003) Risk to breeding success of fish-eating Ardeids due to persistent organic contaminants in Hong Kong: evidence from organochlorine compounds in eggs. Water Res 37:459–467

Department of Environmental Quality, Louisiana, 2003. http://www.deq.louisiana.gov/portal/Portals/0/technology/recap/2003/RECAP%202003%20Appendix%20I%20Tables%2017,18,%20Worksheets%201-8.pdf.

Dong YH, An Q, Gong Z, Wang H (2002) Bioindication of organochlorine pesticides by night heron in Taihu wetland ecosystem. Chin J Appl Ecol 13:209–212

Dong YH, Wang H, An Q, Ruiz X, Fasola M, Zhang YM (2004) Residues of organochlorinated pesticides in eggs of water birds from Tai Lake in China. Environ Geochem Health 26:259–268

FishBase (2013). http://www.fishbase.org/search.php

Furness RW (1993) Birds as monitors of pollutants. In: Furness RW, Greenwood JJD (eds) Birds as Monitors of Environmental Change. Champman Hall, London, UK, pp 86–143

Kong KY, Cheung KC, Wong CKC, Wong MH (2005) The residual dynamic of polycyclic aromatic hydrocarbons and organochlorine pesticides in fishponds of the Pearl River Delta, South China. Water Res 39:1831–1843

Lam JCW, Kajiwara N, Ramu K, Tanabe S, Lam PKS (2007) Assessment of polybrominated diphenyl ethers in eggs of waterbirds from South China. Environ Pollut 148:258–267

Lam JCW, Murphy MB, Wang Y, Tanabe S, Giesy JP, Lam PKS (2008) Risk assessment of organohalogenated compounds in water bird eggs from South China. Environ Sci Technol 42:6296–6302

Leung SF (1997) The population dynamics of *Metapenaeus ensis* (Crustacea: Decapoda: Penaeidae) in a traditional tidal shrimp pond at the Mai Po Marshes Nature Reserve. Hong Kong J Zool 242:77–96

Leung SY, Kwok CK, Nie XP, Cheung KC, Wong MH (2010) Risk assessment of residual DDTs in freshwater and marine fish cultivated around the Pearl River Delta, China. Arch Environ Contam Toxicol 58:415–430

Ley JA (2007) Indo-Pacific Tarpon *Megalops cyprinoids*: a review and ecological assessment. In: Ault JS (ed) Biology and Management of the World Tarpon And Bonefish Fisheries. CRC Press, London

Liang Y, Wong MH (2003) Spatial and temporal organic and heavy metal pollution at Mai Po Marshes Nature Reserve, Hong Kong. Chemosphere 52:1647–1658

Liang Y, Wong MH, Shutes RBE, Revitt DM (1999) Ecological risk assessment of polychlorinated biphenyl contamination in the Mai Po Marshes Nature Reserve, Hong Kong. Water Res 33:1337–1346

Liang Y, Tse MF, Young L, Wong MH (2007) Distribution patterns of polycyclic aromatic hydrocarbons (PAHs) in the sediments and fish at Mai Po Marshes Nature Reserve, Hong Kong. Water Res 41:1303–1311

Mackay D, Shiu WY, Ma KC (1997) Illustrated handbook of physical–chemical properties and environmental fate for organic chemicals. Lewis publishers, Boca Raton

Maruya KA, Lee RF (1998) Biota-sediment accumulation and trophic transfer factors for extremely hydrophobic polychlorinated biphenyls. Environ Toxicol Chem 17:2463–2469

Masterson J (2007) Smithsonian Marine Station at Fort Pierce. http://www.sms.si.edu/irlspec/Oreochromis_mossambicus.htm

Mörner J, Bos R, Fredrix M (2002) Reducing and eliminating the use of Persistent Organic Pesticides: Guidance on alternative strategies for sustainable pest and vector management. United Nations Environment Program, World Health Organization, Food and Agriculture Organization. http://www.chem.unep.ch/pops/pdf/redelipops/redelipops.pdf

Paripatananont T (2002) Snakehead and *Pangasius* catfish. In: Webster CD, Lim CE (eds) Nutrient Requirements and Feeding of Finfish for Aquaculture. CABI, England, pp 396–401

Pereiara MG, Walker LA, Best J, Shore RF (2009) Concentrations of polycyclic aromatic hydrocarbons (PAHs) in the eggs of predatory birds in Britain. Environ Sci Technol 43:9010–9015

Ruan L, Zhang Y, Zhao D, Dong Y, Fasola M (2003) *Egretta garzetta* as a bioindicator of environmental pollution in Tai Lake region. Chin J Appl Ecol 14:263–268

Sang HH, Dong HN, Tae HK (2001) Comparison of the breeding ecology of little egret *Egretta garzetta* and black-crowned night heron *Nycticorax nycticonax*. Korean J Ornithol 8:35–45

Sanpera C, Ruiz X, Jover L, Llorente G, Jabeen R, Muhammad A, Boncompagni E, Fasola M (2003) Persistent organic pollutants in Little Egret eggs from selected wetlands in Pakistan. Arch Environ Contam Toxicol 44:360–368

Simpson CD, Wilcock RJ, Smith TJ, Wilkins AL, Langdon AG (1995) Determination of octanol–water partition coefficients for the major components of technical chlordane. Bull Environ Contam Toxicol 55:149–153

Stokstad E (2007) Species conservation. Can the bald eagle still soar after it is delisted? Science 316:1689–1690

Thomann RV, Komlos J (1999) Model of biota–sediment accumulation factor for polycyclic aromatic hydrocarbons. Environ Toxicol Chem 18:1060–1068

United Nations Environment Program (2005) Ridding the world of POPs: A guide to the Stockholm Convention on persistent organic pollutants. http://www.pops.int/documents/guidance/beg_guide.pdf

United State Environmental Protection Agency (USEPA), (1996a) Method 3540C: Soxhlet extraction. http://www.epa.gov/SW-846/pdfs/3540c.pdf

United State Environmental Protection Agency (USEPA), (1996b) Method 3620C: Florisil cleanup. http://www.epa.gov/SW-846/pdfs/3620c.pdf

United State Environmental Protection Agency (USEPA), (1996c) Method 8270C: Semivolatile Organic Compounds by Gas Chromatography/Mass Spectrometry (GC/MS). http://www.epa.gov/SW-846/pdfs/8270d.pdf

United State Environmental Protection Agency (USEPA), (2000) Bioaccumulation testing and interpretation for the purpose of sediment quality assessment: status and needs. http://www.epa.gov/ost/cs/biotesting/Chem-3.pdf

Van der Oost R, Opperhuizen A, Satumalay K, Heida H, Vermeulen NPE (1996) Biomonitoring of aquatic pollution with feral eel (*Anguilla anguilla*) I: bioaccumulation: biota–sediment ratios of PCBs, OCPs, PCDDs, and PCDFs. Aquat Toxicol 35:21–46

Vidal M, Dominguez J, Luis A (2011) Spatial and temporal patterns of PAHs in birds from northwestern Iberia after a major oil spill. Sci Total Environ 409:2668–2673

Vos JG, Dybing E, Greim HA, Ladefoged O, Lambré C, Tarazona JV et al (2000) Health effects of endocrine-disrupting chemicals on wildlife, with special reference to the European situation. Crit Rev Toxicol 30:71–133

Wong CS, Capel PD, Nowell LH (2001) National-scale, field-based evaluation of the biota-sediment accumulation factor model. Environ Sci Technol 35:1709–1715

Wong MH, Leung AOW, Choi M, Cheung KC (2005) A review on the POP pesticides in China, with emphasis on DDT loadings in human milk. Chemosphere 60:740–752

Wong HL, Giesy JP, Lam PKS (2006) Organochlorine insecticides in mudflats of Hong Kong, China. Arch Environ Contam Toxicol 50:153–165

Young L, Melville DS (1993) The conservation of Deep Bay, Hong Kong. In: Morton B (ed) The Marine Biology of the South China Sea. Hong Kong University Press, Hong Kong, pp 211–232

Zhou HY, Wong MH (2000) Mercury accumulation in freshwater fish with emphasis on the dietary influence. Water Res 34:4234–4242

Zhou HY, Cheung RYH, Cha KM, Wong MH (1998) Metal concentrations in sediments and tilapia collected from inland waters of Hong Kong. Water Res 32:3331–3340

Zhou HY, Cheung RYH, Wong MH (1999) Bioaccumulation of organochlorines in freshwater fish with different feeding modes cultured in treated wastewater. Water Res 33:2747–2756

Supplementary material for:

Biota-Sediment Accumulation Factor (BSAF), Bioaccumulation Factor (BAF), and Contaminant Levels in Prey Fish to Indicate The Extent of PAHs and OCPs Contamination in Eggs of Waterbirds

C.K. Kwok[1], Y. Liang[1*], S.Y. Leung[1], H. Wang[2], Y.H. Dong[2], L. Young[3], J.P. Giesy[4,5,6,7], M.H. Wong[1*]

[1]Croucher Institute for Environmental Sciences, and Department of Biology, Hong Kong Baptist University, Hong Kong SAR, PR China

[2]Institute of Soil Science, Chinese Academy of Sciences (ISSAS), Nanjing, Jiangsu, PR China

[3]World Wide Fund Hong Kong, Mai Po Nature Reserve, Hong Kong SAR, PR China

[4]State Key Laboratory on Marine Pollution Control, City University of Hong Kong, and Xiamen University, PR China

[5]Biomedical Sciences and Toxicology Centre, University of Saskatchewan, Saskatoon, Saskatchewan, Canada

[6]Zoology Department, and Center for Integrative Toxicology, Michigan State University, East Lansing, MI 48824, USA.

[7]Environmental Science Program, Nanjing University, Nanjing, PR China

*Corresponding authors: phone: +852-3411-7746; fax: +852-3411-7743; e-mail: mhwong@hkbu.edu.hk (M.H. Wong); yliang@hkbu.edu.hk (Y. Liang)

Table S1. Concentrations of ∑OCPs in muscles of fish and shrimp collected in Mai Po Ramsar site.

(x 10^3 ng/g, lw)	CC	GM (S)	GM (L)	Tenp (L)	Tarpon (M)	Til (L)	Til (S)	SH (L)	MC (L)	MC (M)	Sh (S)	Sh (M)	Sh (L)	Sh (XL)
Hexachlorobenzene	NA	NA	NA	7.5 [1]	NA	NA	NA	NA	NA	NA	NA	NA	NA	NA
Trans-Chlordane	0.12 [a]	0.11 [a]	0.079 [a]	0.070 [a]	1.7 [b]	1.9 [b]	2.7 [b]	NA	2.2 [b]	NA	NA	2.8 [1]	1.4 [1]	NA
Cis-Chlordane	0.16 [a]	0.16 [a]	0.10 [a]	0.091 [a]	1.7 [bc]	1.8 [bc]	2.3 [c]	0.55 [ab]	2.0 [bc]	0.89 [1]	NA	NA	NA	NA
Trans-Nonachlor	0.18 [a]	0.10 [a]	0.091 [a]	0.073 [a]	2.4 [d]	1.8 [bcd]	2.4 [d]	0.58 [abc]	2.1 [cd]	1.4 [abcd]	NA	NA	NA	0.30 [ab]
p,p'-DDE	1.0 [a]	0.55 [a]	0.61 [a]	1.0 [a]	5.4 [b]	2.0 [a]	2.9 [a]	0.72 [a]	2.4 [a]	1.3 [a]	2.2 [a]	1.3 [a]	2.1 [a]	1.3 [a]
Cis-Nonachlor	0.31 [a]	0.18 [a]	0.17 [a]	0.19 [a]	3.3 [b]	2.6 [b]	3.3 [b]	0.79 [1]	2.6 [b]	NA	NA	NA	NA	NA
ΣOCPs	**1.8 [a]**	**1.1 [a]**	**1.1 [a]**	**1.5 [a]**	**13 [b]**	**10 [b]**	**13 [b]**	**2.2 [a]**	**10 [b]**	**3.0 [a]**	**2.2 [a]**	**1.8 [a]**	**2.5 [a]**	**1.6 [a]**

[1] Statistical analysis was not performed as only one sample was collected. NA: below detection limit.

- For size of fish: small (S): 40-80 g, medium (M): 81-330 g, large (L): 331-770 g. For size of shrimps: small (S): < 6 g; medium (M): 6-10 g, large (L): 11-15 g; extra large (XL): >15 g. CC: Crucian carp; GM: Grey mullet; Tenp: Tenpounder; Tarpon: Indo-Pacific tarpon; Til: Tilapia; SH: Snakehead; MC: Mud carp; Sh: gei wai shrimp (n = 3).
- For each individual OCP, concentrations in a row followed by the same letter are not significantly different at 0.05 probability level according to Duncan test.

Table S2. Concentrations of ∑PAHs in muscles of fish and shrimp collected in Mai Po Ramsar site.

(x 10^3 ng/g, lw)	CC	GM (S)	GM (L)	Tenp (L)	Tarpon	Til (L)	Til (S)	SH (L)	MC (L)	MC (M)	Sh (S)	Sh (M)	Sh (L)	Sh (XL)
Naphthalene	0.69 [a]	0.33 [a]	0.30 [a]	0.33 [a]	13 [bc]	13 [bc]	16 [c]	3.9 [ab]	11 [bc]	5.9 [ab]	10 [bc]	6.4 [ab]	5.9 [ab]	6.1 [ab]
Acenaphtylene	NA	0.10 [1]	NA	NA	NA	NA	NA	NA	NA	NA	NA	NA	NA	NA
Acenaphthene	NA	0.064 [1]	NA	NA	NA	NA	NA	NA	NA	NA	NA	NA	NA	NA
Fluorene	0.14 [a]	0.069 [a]	0.098 [a]	0.083 [a]	1.3 [b]	1.3 [b]	2.5 [c]	0.23 [a]	1.3 [b]	0.60 [ab]	0.98 [ab]	0.62 [ab]	0.64 [ab]	0.67 [ab]
Phenathrene	0.49 [a]	0.32 [a]	0.32 [a]	0.24 [a]	4.1 [abc]	4.9 [bc]	7.3 [c]	1.4 [ab]	5.0 [bc]	3.6 [abc]	4.9 [bc]	3.5 [abc]	2.9 [ab]	3.0 [ab]
Anthracene	NA	0.19	NA	NA	NA	NA	NA	NA	NA	NA	NA	NA	NA	NA
Fluoranthene	0.29 [a]	0.15 [a]	0.17 [a]	0.16 [a]	5.1 [d]	2.9 [abcd]	4.2 [cd]	0.98 [ab]	3.9 [bcd]	1.8 [abc]	2.9 [abcd]	2.5 [abcd]	1.6 [abc]	1.9 [abc]
Pyrene	0.26 [a]	0.14 [a]	0.13 [a]	0.13 [a]	4.0 [ab]	2.0 [a]	3.0 [ab]	0.72 [a]	2.3 [a]	1.2 [a]	3.0 [ab]	6.9 [b]	2.8 [a]	3.1 [ab]
Benz(a)anthracene	NA	NA	NA	NA	NA	0.58 [1]	0.63 [a]	0.15 [1]	0.60 [a]	0.35 [a]	0.39 [a]	0.60 [a]	0.40 [1]	NA
Chrysene	NA	NA	NA	NA	NA	1.4 [1]	1.4 [a]	0.32 [1]	0.93 [a]	0.41 [a]	0.58 [a]	1.1 [a]	0.52 [1]	NA
ΣPAHs	**1.9 [a]**	**1.1 [a]**	**0.98 [a]**	**0.95 [a]**	**27 [bc]**	**24 [bc]**	**35 [c]**	**7.5 [ab]**	**25 [bc]**	**14 [ab]**	**23 [bc]**	**22 [abc]**	**14 [ab]**	**15 [abc]**

Notes are the same as in STable 1.