Toxicity Reference Values for Protecting Aquatic Birds in China from the Effects of Polychlorinated Biphenyls

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1 Introduction

Polychlorinated biphenyls (PCBs) are widely distributed, persistent, bioaccumulative, and toxic pollutants of abiotic matrices, such as soils, sediments, and water, and of wildlife such as fish and birds (Kannan et al. 2000; Giesy et al. 1994b). PCBs are stable in water and adsorb to particles that can be deposited in sediment or accumulated in aquatic food webs. Because of their persistent and lipophilic properties, PCBs bioaccumulate and biomagnify through aquatic food webs, and if thresholds for adverse effects are exceeded, cause effects on wildlife. As top predators that feed at the top of the aquatic food chain, fish-eating birds are exposed to greater concentrations of PCBs (Giesy et al. 1994a; Bosveld and Van den Berg 1994). Potential adverse effects reported for PCBs on wild birds include reduced hatchability, embryonic deformities, immune suppression, mortality (Kannan et al. 2000; Giesy et al. 1994a; Bosveld and Van den Berg 1994; Brunström 1989; Bosveld et al. 1995, 2000; Hoffman et al. 1998), and population-level effects (CCME 1998; Sanderson et al. 1994).

PCBs cause dioxin-like effects by binding to the aryl hydrocarbon receptor (AhR) in birds (Safe 1990, 1994; Kennedy et al. 1996). The primary PCB congeners that cause AhR-mediated effects to birds are the non- and mono-*ortho*-substituted congeners (Bosveld and Van den Berg 1994; Bosveld et al. 2000). As the most sensitive biological effect, ethoxyresorufin-*O*-deethylase (EROD) activity induction has been suggested to be a suitable biochemical indicator for exposure to dioxin-like compounds, such as some PCB congeners (Elliott et al. 2001). It is thought that the developing embryo is the life stage that is most sensitive to the toxic effects of pollutants (Lam et al. 2008; Peterson et al. 1993). Accordingly, PCB concentrations in eggs were the more predictive measure of exposure to derive toxic reference values (TRVs). The tissue-based risk of great blue herons was assessed on the basis of an egg-based TRV, and was developed by taking the geometric mean of effect concentrations in three egg-injection studies (Seston et al. 2010).

PCB congeners have different toxic potencies because their physical and chemical properties and structures are different. Such differences determine the binding affinity of PCBs to the AhR. Assessments of the PCB hazard to humans and wildlife are complicated because PCBs occur in mixtures that change as a function of time from weathering, differential accumulation, and metabolism. Log-K_{ow} values for the PCBs are between 4.3 and 8.26. PCBs that have even moderate K_{ow} values accumulate in aquatic organisms and their predators. Relative Potency Factors (RePs) have been used to calculate concentrations of 2,3,7,8-TCDD toxicity equivalents (TEQ) in samples as the sum of the product of the ReP multiplied by the concentration of the respective congeners (Van den Berg et al. 1998).

Wildlife, such as birds, consumes persistent substances mainly via consumption of fish, crustaceans, invertebrates, and plants. Governments have established allowable residue concentrations in tissues to protect wildlife that feed on aquatic organisms contaminated by persistent, bioaccumulative, and toxic substances. Tissue residue guidelines (TRGs) are concentrations of xenobiotics in tissues that are established for aquatic biota to protect wildlife that consume them. Generally, as concentrations of xenobiotics increase, the greater is the expectation that adverse effects will occur on birds and mammals feeding on aquatic organisms (CCME 1998). The USEPA (1995a) used NOAEL (no observed adverse effects levels) or LOAEL (lowest observed adverse effects levels) values to derive wildlife criteria for PCBs. Hatching success of pheasant eggs was the endpoint and appropriate uncertainty factors were used. In addition, the EPA derived wildlife criteria for the PCBs for kingfisher, silver gulls, and bald eagles, and the geometric mean of the values for these three species was taken as the PCB wildlife criterion for birds by the USEPA. The tissue residue guideline for PCBs in aquatic birds derived in Canada was 2.4 ng TEQs/kg food wet mass (wm), and was based on a toxicity study in white Leghorn hens (CCME 1998, 2001).

We had two objectives in the present study:

1. Derive TRVs and TRGs for the effects of PCBs on aquatic birds by using the toxicity percentile rank method (TPRM), the species sensitivity distribution (SSD) and critical study approach (CSA), along with the method used in the USA and Canada for deriving the wildlife criteria for PCBs in birds.
2. Assess the PCB hazard to birds by comparing TRVs and TRGs derived in this study, with the actual concentrations of PCBs measured in birds and fish in selected regions of China. The additional value contributed by this study is that it provides a scientific baseline for risk management of PCBs in China.

2 Data Collection and Analysis Methods

2.1 Selection of Representative Species in China and Toxicity Data

The primary criterion for selecting representative avian species is their exposure to pollutants via aquatic food webs (USEPA 1995a), such as fish-eating birds. The night heron (*Nycticorax nycticorax*), little egret (*Egretta garzetta*), and Eurasian spoonbill (*Platalea leucorodia*) were selected as three representative avian species in China (ZRQ). All of these are widely distributed in Chinese aquatic ecosystems and are known to feed on aquatic prey (Barter et al. 2005). These three species have been studied extensively as bioindicators of wetland health and environmental pollution (Lam et al. 2008; Levengood et al. 2007; An et al. 2006). Body masses (bm) and rates of food ingestion (FI) for these three avian species are shown in Table 1.

Table 1 Body masses (bm) and food ingestion (FI) rates of several avian birds

Avian species	bm (kg)	FI (kg/day wm[a])	FI:bm	Reference
Night heron	0.706	0.239	0.34	Zhang et al. (2013)
Little egret	0.342	0.148	0.43	Zhang et al. (2013)
Eurasian spoonbill	2.232	0.514	0.23	Zhang et al. (2013)
Common tern	0.127	0.0774	0.61	Nagy (2001)
Chicken	2.0	0.134	0.067	USEPA (1995a)
Ring-necked pheasant	1.1	0.0638	0.058	USEPA (1995a)
Japanese quail	0.12	0.012	0.10	USEPA (1995a)
Northern bobwhite	0.04	0.0072	0.18	USEPA (2007)
Mourning dove	0.128	0.058[b]	0.45	Nelson and Martin (1953)
Ring dove	0.149	0.0169	0.11	USEPA (2007)
American kestrel	0.12	0.0444	0.37	USEPA (1995a)
Screech owl	0.194	0.02	0.10	USEPA (2007)
Mallard	1.082	0.25	0.23	CCME (1998)

[a] wm stands for wet mass

[b] Calculated from the allometric equation (Nagy 2001): $FI = 2.065 \times bm^{0.689}$

The effects of PCBs on birds have been reviewed and summarized (USEPA 1995a; Barron et al. 1995; Bosveld and Van den Berg 1994), and toxicity threshold values for PCBs have been derived from NOAEL or LOAEL levels established for several toxicity endpoints. Toxicity data for dietary exposure were converted to tolerable daily intake (TDI) values, which were calculated from bm and food ingestion rates of selected surrogate birds. Utilizable NOAEL or LOAEL values were selected, based on the principles given in the following document: "Protocol for the derivation of Canadian tissue residue guidelines for the protection of wildlife that consume aquatic biota" (CCME 1998). The main principals followed are as follows: (1) studies were constructed under suitable control conditions and considered ecological-relevant endpoints, such as reproduction and embryonic development; (2) only chronic or subchronic studies with a clear dose–response relationship were accepted; (3) the form and dosage of tested chemicals were reported in the study.

2.2 *Methods for Deriving TRVs and TRGs*

PCBs occur in the environment as weathered mixtures, and weathered residue profiles differ from those of the original technical mixtures. Therefore, assessment of hazards posed by PCBs to wildlife must account for changes in the relative proportions of PCB congeners and their different toxic potencies. Accordingly, the concept of Relative Potency Factors (ReP) was introduced to allow comparisons of the toxicity of a compound relative to TCDD, based on its available in vivo and in vitro data (Van den Berg et al. 1998). In this approach, it is assumed that the combined effects of different congeners were either dose- or concentration-additive. Concentrations of 2,3,7,8-TCDD equivalents (TEQs) of PCBs can be calculated by using toxic equivalency factors (TEFs) and available chemical residue data (1). Application of an NOAEL or LOAEL value as a reference dose could either be overprotective or under-protective and may not reflect the specific dose–response relationship (Kannan et al. 2000). To address this problem, the geometric mean of the NOAEL and LOAEL values are used as the reference concentration (RC) (Kannan et al. 2000) (2). If the NOAEL value was not determined in a particular study, it can be estimated by dividing the LOAEL by a factor of 5.6 (CCME 1998) (3). The tolerable daily intake (TDI) is calculated as shown in (4).

$$\mathrm{TEQ} = \sum \left(\mathrm{PCB_i} \times \mathrm{TEF_i}\right) \tag{1}$$

$$\mathrm{RC} = \left(\mathrm{NOAEL} \times \mathrm{LOAEL}\right)^{0.5} \tag{2}$$

$$\mathrm{NOAEL} = \mathrm{LOAEL} / 5.6 \tag{3}$$

$$\mathrm{TDI} = \mathrm{RC} \times \left(\mathrm{FI} / \mathrm{bm}\right) \tag{4}$$

Three methods used to derive TRGs and TRVs are (1) Species sensitivity distribution (SSD), (2) Critical study approach (CSA), and (3) Toxicity percentile rank method (TPRM). Each of the three has advantages and disadvantages. A species sensitivity distribution (SSD) is a probability distribution function that can be used to describe the range of tolerances among species (Leo Posthuma et al. 2002). The SSD method has been used widely in aquatic ecological risk assessment and derivation of water quality criteria (WQC) for aquatic biota (Caldwell et al. 2008; Hall et al. 2009). The SSD makes full use of available toxicity data and represents the whole ecosystem. But this approach is not often applied when assessing risks to wildlife, because so little toxicity data for wildlife are available. We used the SSD method in this study to derive the TRVs and TRGs of PCBs for protection of fish-eating birds, and we used the most sensitive endpoint data for each species (USEPA 2005). The SSD approach assumes that sensitivities of species can be described by a specified statistical distribution (e.g., normal distribution). If the selected toxic data for PCBs can be described by using a log-normal distribution, then the ETX2.0 program can be employed to fit the distribution. Calculating an HC_5 (Hazard Concentration affecting 5% of species) via this program gives a value that protects 95% species from contaminants. Moreover, it provides two-sided 90% confidence limits designated as an upper limit (UL HC_5) and lower limit (LL HC_5) (Zhang et al. 2013).

The critical study approach (CSA) is a primary method used for risk assessment and criteria derivation for wildlife (Kannan et al. 2000; CCME 2001; USEPA 1995a; Sample et al. 1993; Newsted et al. 2005). The CSA method has the advantage of requiring less data and being simpler to calculate. This method depends mainly on the toxicity values of sensitive species and has greater uncertainty. Results from available toxicity studies on the targeted species were selected in this method as the basis for deriving TRVs (Blankenship et al. 2008). TRVs for wildlife were then calculated by using the lowest toxicity value from the critical study (tissue level or dietary concentration); appropriate uncertainty factors (UFs) were also applied.

Uncertainty factors were determined primarily from guidance given by the US-EPA (USEPA 1995a, b; Weseloh et al. 1995). Three types of uncertainty factors were considered: interspecies uncertainty factor (UF_A), sub-chronic to chronic uncertainty factor (UF_S), and LOAEL-to-NOAEL uncertainty factor (UF_L). Values of 1–10 were assigned to represent the degree of uncertainty for each factor and were based on the nature of available scientific information as well as professional judgment.

The toxicity percentile rank method (TPRM) is the standard method recommended by the USEPA for deriving water quality criteria for protecting aquatic organisms (USEPA 1985). The TPRM more comprehensively reflects the toxic effects of pollutants to organisms and ultimately provides better protection for wildlife. When using the TPRM, the reference concentrations (RC) for avian species are first ordered from largest to least, and ranks (*R*) are assigned to RCs from 1 for the lowest to *N* (*N* is the number of avian species) for the highest. Second, the cumulative probability *P* is calculated for each species using the equation: $P = R/(N+1)$. Finally, four RCs, which have cumulative probabilities closest to 0.05 (always the four least RCs,) are selected as the basis to calculate the TRVs (5–8).

$$S^2 = \frac{\sum\left[(\ln \mathrm{RC})^2\right] - \left[\sum(\ln \mathrm{RC})\right]^2/4}{\sum(\mathrm{P}) - \left[\sum\left(\sqrt{\mathrm{P}}\right)\right]^2/4} \tag{5}$$

$$L = \left\{\sum(\ln \mathrm{RC}) - S\left[\sum\left(\sqrt{\mathrm{P}}\right)\right]\right\}/4 \tag{6}$$

$$A = S\left(\sqrt{0.05}\right) + L \tag{7}$$

$$\mathrm{TRV} = e^{A} \tag{8}$$

3 Review of PCB Bird Toxicity Studies

The toxicity of PCBs to birds, emphasizing reproduction and developmental effects, was summarized by Barron et al. (1995). To augment the information from Barron et al. (1995), additional recent and relevant toxicity studies of PCB effects on birds were compiled, reviewed, and critiqued. All available toxicity data (both diet and tissue data) were summarized and are presented in Table 2.

3.1 Domestic Chicken (Gallus gallus domesticus)

It has been shown in several studies that chickens are among the most sensitive species to the effects of PCBs; moreover, PCB126 was the most toxic congener and

Table 2 A summary of the toxicity of PCB isomers and mixtures to birds (both for tissue and diet)

Species	PCBs	Toxicity end point	NOAEL[a]	LOAEL[b]	References
Chicken (tissue)	PCB126	Egg mortality		0.2 ng/g wm	Powell et al. (1996)
	PCB126	EROD[c] activity		0.3 ng/g wm	Hoffman et al. (1998)
	PCB77	EROD activity	0.12 ng/g wm	1.2 ng/g wm	Hoffman et al. (1998)
	PCB126	Egg mortality		1.0 ng/g wm	McKernan et al. (2007)
	PCB126	Thymocyte apoptosis	0.13 ng/g wm	0.32 ng/g wm	Goff et al. (2005)
	PCB126	Immune function		0.25 ng/g egg	Lavoie and Grasman (2007)
	PCB126	Egg mortality	0.051 ng/g wm	0.128 ng/g wm	Fox and Grasman (1999)
	PCB1254, 1242	MDI[d] activity		6.7 mg/kg wm	Gould et al. (1999)
	PCBs	Hatching success	0.95 mg/kg wm	1.5 mg/kg wm	Britton and Huston (1973)
		Hatching success	0.36 mg/kg wm	2.5 mg/kg wm	Scott (1977)
		Egg production		5 mg/kg wm	Platonow and Reinhart (1973)
		Hatching success		4 mg/kg wm	Tumasonis et al. (1973)
Chicken (diet)	Aroclor 1016, 1221, 1254	Reproductive efficiency	20 mg/kg food		Lillie et al. (1974, 1975)
	Aroclor 1232, 1268	Egg production		20 mg/kg food	Lillie et al. (1974)
	Aroclor 1232, 1242, 1248	Hatching success	5 mg/kg food	10 mg/kg food	Britton and Huston (1973), Lillie et al. (1975)
	Aroclor 1242	Hatching success	2 mg/kg food	20 mg/kg food	Lillie et al. (1974)
	Aroclor 1248, 1254	Chick growth		2 mg/kg food	Lillie et al. (1974)
	Aroclor 1248	Hatching success	1 mg/kg food	10 mg/kg food	Scott (1977)
	Aroclor 1254	Egg production		5 mg/kg food	Platonow and Reinhart (1973)
	Aroclor 1254	Hatching success		50 mg/kg food	Tumasonis et al. (1973)
Double-crested cormorant (tissue)	PCB126	Egg mortality		25 ng/g wm	Powell et al. (1997)
	PCB126	EROD induction		70 ng/g wm	Powell et al. (1998)
	PCBs	Egg mortality		3.5 mg/kg wm	Tillitt et al. (1992)

(continued)

Table 2 (continued)

Species	PCBs	Toxicity end point	NOAEL[a]	LOAEL[b]	References
American kestrel (tissue)	PCB126	EROD induction	23 ng/g wm	233 ng/g wm	Hoffman et al. (1998)
	PCB77	EROD induction	100 ng/g wm	1000 ng/g wm	Hoffman et al. (1998)
	PCB126	EROD induction		50 ng/g wm	Hoffman et al. (1996)
American kestrel (diet)	Aroclor 1254	Male fertility		33 mg/kg food	Bird et al. (1983)
	Aroclor 1254	Female fertility	0.5 mg/kg food	5 mg/kg food	Linger and Peakall (1970)
Common tern (tissue)	PCBs	EROD induction	25ngTEQ[e]s/g lipid		Bosveld et al. (2000)
	PCB126	EROD induction		44 ng/g wm	Hoffman et al. (1998)
	PCBs	Reproductive success	7 mg/kg wm	8 mg/kg wm	Bosveld and Van den Berg (1994)
		Reproductive success	5 mg/kg wm		Hoffman et al. (1993)
		Reproductive success	4 ng TEQs/g lipid		Bosveld and Van den Berg (1994)
Common tern (diet)	PCBs	EROD induction	0.6 ng TEQs/g food		Bosveld et al. (2000)
Caspian tern (tissue)	PCBs	Reproductive success		4.2 mg/kg wm	Yamashita et al. (1993)
	PCBs	Egg shell thickness	30 mg/kg wm		Struger and Weseloh (1985)
Bald eagle (tissue)	PCBs	EROD induction	0.1 ng TEQs/g wm	0.21 ng TEQs/g wm	Elliott John et al. (1996)
	PCBs	Reproductive success		4 mg/kg wm	Wiemeyer et al. (1984)
		Reproductive success		13 mg/kg wm	Bosveld and Van den Berg (1994)
Osprey (tissue)	PCBs	EROD induction	0.037 ng TEQs/g wm	0.13 ng TEQs/g wm	Elliott et al. (2001)
Herring gull (tissue)	PCBs	Reproductive success		5 mg/kg wm	Ludwig et al. (1993)
Great horned owl (tissue)	PCBs	EROD induction	0.14 ng TEQs/g wm	0.4 ng TEQs/g wm	Strause et al. (2007)
Great blue heron (tissue)	PCBs	Reproductive success	7.8 mg/kg wm		Boily et al. (1994)

Black-crowned night heron (tissue)	PCBs	Breeding success	10.9 mg/kg wm		Tremblay and Ellison (1980)
Mallard (tissue)	Aroclor 1254	Reproductive success	23.3 mg/kg wm		Custer and Heinz (1980)
	Aroclor 1242	Egg shell thinning		105 mg/kg wm	Haseltine and Prouty (1980)
Mallard (diet)	Aroclor 1254	Reproductive success	25 mg/kg food		Custer and Heinz (1980)
	Aroclor 1242	Egg shell thing		150 mg/kg food	Haseltine and Prouty (1980)
Screech owl (tissue)	Aroclor 1248	Reproductive success	7.1 mg/kg wm		Anne et al. (1980)
Screech owl (diet)	Aroclor 1248	Reproductive success	3 mg/kg food		Anne et al. (1980)
Forster's tern (tissue)	PCBs	Reproductive success	7 mg/kg wm	19 mg/kg wm	Bosveld and Van den Berg (1994)
	PCBs	Hatching success	4.5 mg/kg wm	22.2 mg/kg wm	Kubiak et al. (1989)
		Hatching success	0.2 ng TEQs/g wm	2.2 ng TEQs/g wm	Kubiak et al. (1989)
Ringed turtle dove (tissue)	Aroclor 1254	Hatching success		16 mg/kg wm	Peakall et al. (1972)
Ringed turtle dove (diet)	Aroclor 1254	Hatching success		10 mg/kg food	Peakall et al. (1972), Peakall and Peakall (1973)
	Aroclor 1254	Brain neurotransmitter concentrations	1 mg/kg food	10 mg/kg food	Heinz et al. (1980)
Mourning dove (diet)	Aroclor 1254	Reproductive behavior		10 mg/kg food	Tori and Peterle (1983)
Northern bobwhite (diet)	Aroclor 1254	Reproductive effects	50 mg/kg food		Eisler (1986)
Japanese quail (diet)	Aroclor 1254	Egg shell thickness	50 mg/kg food		Chang and Stokstad (1975)
	Aroclor 1248	Egg shell quality	20 mg/kg food		Scott (1977)
Ring-necked pheasant (diet)	Aroclor 1254	Female fertility		50 mg/kg food	Roberts et al. (1978)

[a] No observed adverse effects level
[b] Lowest observed adverse effects level
[c] Ethoxyresorufin-*O*-deethylase
[d] Monodeiodinase
[e] Toxicity equivalents

the major contributor to TEQs (Wiesmüller et al. 1999; Senthilkumar et al. 2002; Strause et al. 2007).

The toxic effects of PCB126 and PCB77 on chickens (*Gallus gallus*) through hatching were studied, focusing on embryonic development and induction of EROD activity (Hoffman et al. 1998). Doses of two congeners were injected into chicken embryos at the following doses: PCB126 (0.3, 0.5, 1, or 3.2 ng/g wet mass (wm)), or PCB77 (0.12, 1.2, 6, or 12 ng/g wm). The LD_{50} and LOAEL values of PCB126 for chicken were 0.4 and 0.3 ng/g wm and the LD_{50}, NOAEL, and LOAEL of PCB77 were 2.6, 0.12, and 1.2 ng/g wm, respectively. In addition, the effects of PCB126 (0.1, 0.2, 0.4, 0.8, 1.6, 3.2, 6.4, or 12.8 ng PCB126/g wm egg) on hatching and development of chicken were also investigated via injection into eggs. The LOAEL of PCB126 was 0.2 ng/g wm (Powell et al. 1996). Three doses (viz., 0.5, 1, and 2.0 ng PCB126/g wm egg) were injected into eggs, and the LOAEL value, based on hatching success, survival, and edema, was 1.0 ng PCB126/g wm egg (McKernan et al. 2007). The toxic effects of PCB126 on chicken embryos were determined by injection of 0.051, 0.13, 0.32, or 0.80 ng PCB126/g egg, wm. The NOEC and LOEC values (0.051 and 0.13 ng/g wm egg, respectively) for PCB126 (injected into chicken embryos) were based on mortality, immune organ quality, and lymphocyte structure (Fox and Grasman 1999).

The effects of PCB126 on thymus atrophy in chicken embryo was examined by egg injection (0.05, 0.13, 0.32, 0.64, or 0.80 ng/g wm egg); the resulting LD_{50}, NOEC, and LOEC values were respectively 1.01, 0.13, or 0.32 ng/g wm egg (Goff et al. 2005). The effects of PCB126 on death and immune function of chickens were determined by egg injection, and the LOAEL was 0.25 ng/g wm egg (Lavoie and Grasman 2007). LOAELs of total PCBs for reproductive success of chicken were 1,500–5,000 ng/g of egg wm (Barron et al. 1995), which were greater than PCB126 values (the most toxic congener of PCBs).

*3.2 Double-Crested Cormorant (*Phalacrocorax auritus*)*

The double-crested cormorant is a typical fish-eating bird that has exhibited adverse effects attributed to PCB exposure. Such effects include embryonic lethal and developmental defects, from ingesting fish contaminated with PCBs and other substances. Based on a toxic endpoint of egg mortality, the LOAEL value for PCBs was 3,500 ng/g wm egg (Tillitt et al. 1992; Yamashita et al. 1993). In another study, eggs of double-crested cormorants were injected with 5, 10, 25, 50, 100, 200, 400, or 800 ng PCB126/g wm to examine double-crested cormorant toxicity. The LD_{50} and LOAEL values of PCB126 for double-crested cormorant were 158 and 25 ng/g egg, respectively (Powell et al. 1997). The LD_{50} of PCB126 was greater than that for chicken, which value was 2.3 ng/g wm egg. This result indicated that chickens were more sensitive to the effects of PCB126 than were double-crested cormorants (Powell et al. 1997; Fox and Grasman 1999). In another study, the co-planar PCB congener, PCB126, was injected into the eggs of double-crested cormorants at doses of 60, 150, 300, or 600 ng/g egg. Based on induction of EROD activity, the LOAEL and LD_{50} values of PCB126 for double-crested cormorant were 70 and 177 ng/g wm egg, respectively (Powell et al. 1998).

3.3 *Common Tern (***Sterna hirundo***)*

As fish-eating birds that top the aquatic food chain, common terns are exposed to the lipophilic and persistent PCBs (Bosveld et al. 1995). The toxicity of PCB126 on common tern was examined by egg injection at three doses (0, 240 or 434 ng/g wm). The LD_{50} and LOAEL, based on reproductive success and induction of EROD activity, were 104 and 44 ng/g wm, respectively (Hoffman et al. 1998).

Biochemical and reproductive effects of PCB126 on chicks of common terns fed fish containing PCB126, or mixtures of PCB126 and PCB153, were examined (Bosveld et al. 2000). The most sensitive parameter affected by PCBs was induction of EROD activity. A nonlinear concentration–effect relationship was observed between TEQ concentrations and EROD activity induction. The LOAEL value for induction of EROD activity was 25 ng TEQs/g liver lipid mass (lm), and was caused by 0.6 ng TEQs/g wm food. The lipid content of common tern was assumed to be 1.9% (Ricklefs 1979), and the LOAEL value was calculated to be 475 pg TEQs/g wm. Based on reproductive success, the NOAEL of PCBs for common tern was <4 ng TEQs/g lm (Bosveld and Van den Berg 1994).

3.4 *Osprey (***Pandion haliaetus***)*

As a top predator, osprey accumulates lipophilic substances and accordingly can be used as a biological indicator of exposure to PCBs in the aquatic ecosystems (Elliott et al. 2001). Elliott et al. (2001) studied the ecological effects of PCBs on osprey chicks and found that the NOAEL and LOAEL values, based on induction of EROD activity, were 37 and 130 ng TEQs/kg wm, respectively (Elliott et al. 2001).

3.5 *Bald Eagle (***Haliaeetus leucocephalus***)*

As a top predator of the aquatic food chain, bald eagles feed mainly on fish and other fish-eating birds (Knight et al. 1990). PCBs can reduce the reproductive success of bald eagle populations (Anthony et al. 1993; Wiemeyer et al. 1993). Chicks of bald eagles were more sensitive to the effects of PCBs than were adults. Induction of EROD activity was the most sensitive biomarker for bald eagles exposed to PCBs (Sanderson et al. 1994). Based on induction of EROD activity, the LOAEL and NOAEL values were 100 and 210 ng TEQs/kg wm, respectively (Elliott John et al. 1996).

3.6 *American Kestrel (***Falco sparverius***)*

Exposure of the American kestrel to a daily intake of 7 mg PCB/kg bm/day produced a body concentration of 34.1 mg PCBs/kg wm, which was consistent with PCB concentrations in birds collected from the Great Lakes basin (Fernie et al. 2001b; Fisher et al. 2006). Breeding behavior was affected and reproductive success was reduced. Subsequent studies have shown that embryos exposed to PCBs could affect propagation of offspring (Fernie et al. 2001a). The developmental toxicity of American kestrel was studied by egg injection of PCB126 (0, 2.3, 23, or 233 ng/g wm) or PCB77 (0, 100, or 1,000 ng/g wm) (Hoffman et al. 1998). Results showed that the LOAELs, based on induction of EROD activity, were 233 and 1,000 ng/g wm for PCB126 and PCB77, respectively. When American kestrel were exposed orally to 50, 250, or 1,000 ng/g bm, the LOAEL value, based on developmental toxicity, was 50 ng PCB126/g bm (Hoffman et al. 1996).

3.7 *Great Horned Owl (***Bubo virginianus***)*

Great horned owls are another species that feeds at the top of the terrestrial food chain, and hence are very sensitive to PCBs, and are often used as a biological indicator species. Based on induction of EROD activity, the NOAEL and LOAEL values for PCB exposure were 135 and 400 pg TEQs/g wm egg, respectively (Strause et al. 2007; Elliott John et al. 1996).

4 Derivation of TRVs and TRGs

TRGs and TRVs for birds were derived for PCBs by using three approaches: SSD, TPRM, and CSA. The toxic endpoints recorded for PCBs on birds are shown in Table 2. Because of the toxicity differences among PCB congeners, TEQs were selected and used to derive TRVs and TRGs. NOAEL and LOAEL values for the most sensitive toxicity endpoints were selected and the geometric mean of these two values was used as the reference concentration (RC). If a value was available only for the NOAEL or the LOAEL, then the other one was calculated by using (3). Toxicity data were transformed to equivalent concentrations by using TEFs for birds (Van den Berg et al. 1998), and TEFs for PCB commercial mixtures (Table 3).

4.1 *Species Sensitivity Distribution Method*

Using the data selection principles mentioned above, toxicity data (tissue) on PCBs was selected to derive a TRV of PCBs for the following birds: chicken,

Table 3 Toxic equivalent conversion factors for some commercial PCB mixtures for birds (CCME 2001)

Mixture	Conversion factor (ng TEQ/mg product)
Aroclor 1242	234.6
Aroclor 1248	251.3
Aroclor 1254	44.5
Aroclor 1260	25.5

Table 4 Reference concentrations (RC) used to construct SSD curves (pg TEQs/g wm)

Avian species	PCBs[a] NOAEL/LOAEL	TEQs[b] NOAEL/LOAEL	RC[c]	Reference
Chicken	0.051/0.128	0.0051/0.0128	8	Fox and Grasman (1999)
Double-crested cormorant	0.45/25	0.45/2.5	1,060	Powell et al. (1997)
American kestrel	8.9/50	0.89/5	2,110	Hoffman et al. (1996)
Common tern	7.9/44	0.79/4.4	1,860	Hoffman et al. (1998)
Bald eagle	N/N	0.1/0.21	140	Elliott John et al. (1996)
Osprey	N/N	0.037/0.13	70	Elliott et al. (2001)
Great horned owl	N/N	0.14/0.4	240	Strause et al. (2007)
Mallard	23,300/130,480	1.04/5.82	2,460	Custer and Heinz (1980)
Screech owl	7,100/39,760	1.78/9.97	4,210	Anne et al. (1980)
Forster's tern	4,500/22,200	0.2/2.2	660	Kubiak et al. (1989)
Ringed turtle dove	2,857/16,000	0.13/0.71	90	Peakall et al. (1972)

N no data
[a] ng PCBs/g egg
[b] ng TEQs/g wm egg
[c] pg TEQs/g bm/day

double-crested cormorant, American kestrel, common tern, bald eagle, osprey, great horned owl, mallard, screech owl, Forster's tern, and ringed turtle dove (Table 4). From the data on these species, the SSD was constructed and the hazard concentration affecting 5% of species (HC_5) was estimated by using the ETX2.0 software. The HC_5 value, which theoretically protects 95% species, is known to be a good predictor of the threshold for community-level effects. We determined the HC_5 value, along with the 90% upper and lower confidence limits (UL HC_5 and LL HC_5) and show them in Fig. 1. The HC_5 was predicted to be 15.5 pg TEQs/g, wm, which was defined as the TRV with UL HC_5 and LL HC_5 of 1.8 and 54.5 pg TEQs/g, wm, respectively.

Toxicity data on the PCBs for common tern, chicken, ring-necked pheasant, Japanese quail, Northern bobwhite, mourning dove, ringed turtle dove, American kestrel, screech owl, and mallard based on diet were selected to derive the TRG of PCBs (Table 5). Data expressed as TEQs were obtained by use of TEFs (1). The Total Daily Intake (TDI) was calculated from FI and bm using (2) and (4) (Table 1). TDI values were calculated for all ten avian species as shown in Table 5.

The TDI values were then used to construct a SSD curve (Fig. 2). The HC_5 value was predicted to be 3.43 pg TEQs/kg bm/day, and the UL HC_5 and LL HC_5 were

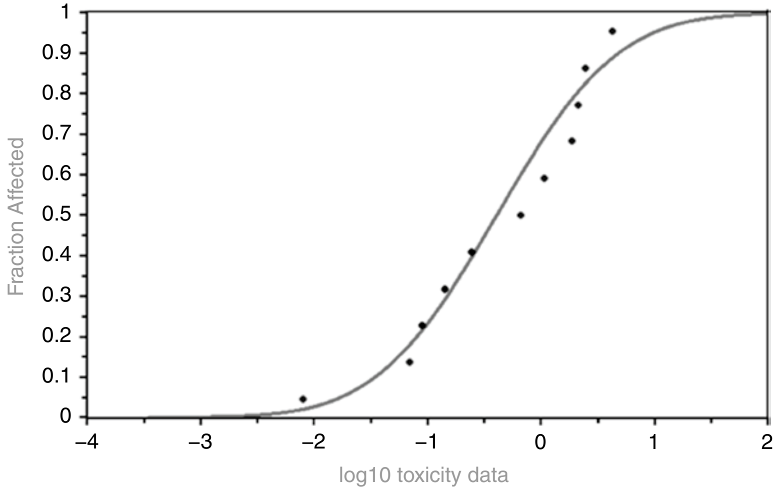

Fig. 1 Distribution of species sensitivity (SSD) for toxicity of PCBs to birds (pg TEQs/g wm). The HC_5 was 15.5 pg TEQs/g wm, and UL HC_5 and LL HC_5 were 1.8 and 54.5 pg TEQs/g wm, respectively

Table 5 The toxicity data values, based on dietary exposure, that were used to fit the SSD curve

Avian species	PCBs[a] NOAEL/ LOAEL	TEQs[b] NOAEL/ LOAEL	TDI[c]	Reference
Common tern	N/N	0.11/0.6	158.6	Bosveld et al. (2000)
Chicken	0.36/2	0.016/0.089	2.6	Lillie et al. (1974)
Ring-necked pheasant	8.9/50	0.4/2.2	54.5	Roberts et al. (1978)
Japanese quail	50/280	2.2/12.3	520	Chang and Stokstad (1975)
Northern bobwhite	50/280	2.2/12.3	936	Eisler (1986)
Mourning dove	1.8/10	0.08/0.45	85.5	Tori and Peterle (1983)
Ringed turtle dove	1/10	0.045/0.45	154	Heinz et al. (1980)
American kestrel	0.5/5	0.022/0.22	25.9	Linger and Peakall (1970)
Screech owl	3/16.8	0.13/0.73	9.5	Anne et al. (1980)
Mallard	25/140	1.1/6.2	598	Custer and Heinz (1980)

TDI tolerable daily intake, *N* no data
[a] mg PCBs/kg wm food
[b] ng TEQs/g wm food
[c] pg TEQs/g bm/day

respectively 0.35 and 12.5 pg TEQs/kg bm/day. Using FI/bm values of three representative avian species listed in Table 1, RCs for these three bird species were calculated to be 10.1, 7.98, and 14.9 pg TEQs/g food. The geometric mean of these three RCs was 10.7 pg TEQs/g food, which was defined as the TRG value in birds for PCBs.

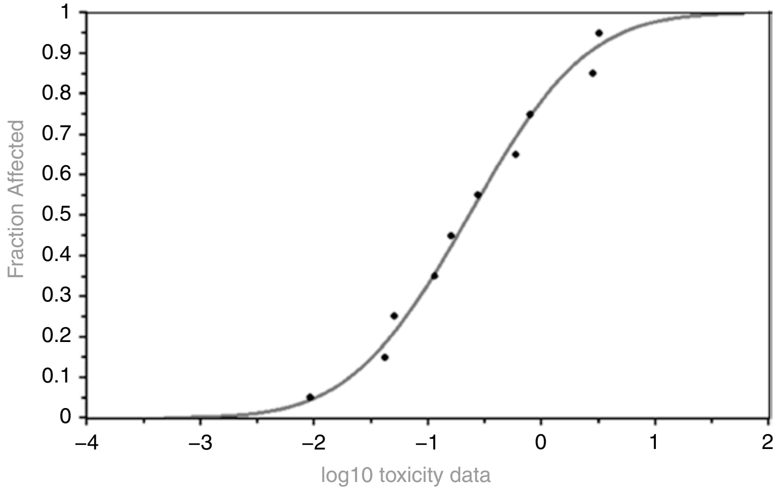

Fig. 2 Distribution of species sensitivity for avian toxicity data of PCBs based on diet exposure. The HC_5 was 3.43 pg TEQs/g/day, and UL HC_5 and LL HC_5 were 0.35 and 12.5 pg TEQs/g bm/day, respectively

4.2 Critical Study Approach

Based on induction of EROD activity as the toxicity endpoint, the NOAEL and LOAEL values for PCBs on osprey were 37 and 130 pg TEQs/g wm, respectively (Elliott et al. 2001). The geometric mean of these two values (69.4 pg TEQs/g wm) was taken as the RC. Similarly, based on induction of EROD activity the NOAEL and LOAEL values for PCBs on bald eagle were 100 and 210 pg TEQs/g wm, respectively (Elliott John et al. 1996), and the RC was 144.9 pg TEQs/g wm. Osprey and bald eagle are two representative birds at the top of the aquatic food web, and both of these species are sensitive to the effects of PCBs. Induction of EROD activity is the most sensitive biological effect, and is a suitable biochemical indicator for exposure to PCBs in birds (Bosveld and Van den Berg 1994; Bosveld et al. 2000). EROD induction activity is the critical endpoint that occurs at the least exposure concentration. Although conservative, application of this assessment endpoint should be protective of population-level adverse effects. By using the toxicity data available for osprey and bald eagle, the TRV for PCB bird effects can be derived for protecting aquatic birds (CCME 1998; Newsted et al. 2005). The UF_A, UF_L, and UF_S were set to be 2, 1, and 3, respectively, and the total UF was 6. The TRV of PCBs for birds was then calculated to be 16.7 pg TEQs/g wm, by dividing the geometric mean of two RCs for these birds by a total uncertainty factor of 6.

The common tern is a piscivorous bird at the top of aquatic food chain that can accumulate PCBs in their tissues and eggs. Based on induction of EROD activity as the toxicity endpoint, the LOAEL value for PCBs on common tern was 0.6 ng

Table 6 The RCs and relevant values used to calculate TRV[a] for TPRM[b] (pg TEQs/g)

Rank	Avian species	RC	ln RC	(ln RC)^2	$P=R/(N+1)$	$P^{0.5}$
4	Bald eagle	140	4.94	24.42	0.33	0.58
3	Ringed turtle dove	90	4.50	20.25	0.25	0.50
2	Osprey	70	4.25	18.05	0.17	0.41
1	Chicken	8	2.08	4.32	0.08	0.29
Sum			15.77	67.04	0.83	1.77

[a]Toxic reference value
[b]Toxicity percentile rank method

Table 7 The TDIs and associated relevant values used to calculate TRG[a] for TPRM (pg TEQs/kg/day)

Rank	Avian species	TDI	ln TDI	(ln TDI)^2	$P=R/(N+1)$	$P^{0.5}$
4	Ring-necked pheasant	54.5	4.00	16.00	0.36	0.60
3	American kestrel	25.9	3.25	10.59	0.27	0.52
2	Screech owl	9.50	2.25	5.07	0.18	0.43
1	Chicken	2.60	0.96	0.91	0.09	0.30
Sum			10.46	32.57	0.91	1.85

[a]Tissue residue guideline

TEQs/g wm (Bosveld et al. 2000). This study was taken as the critical study for deriving a TRG value for PCBs in birds. By employing a total uncertain factor of 6, the calculated TRG value was 42.3 pg/g wm food.

4.3 Toxicity Percentile Rank Method

Four of the lowest RC values for bald eagle, ringed turtle dove, osprey and chicken were selected to calculate the TRV for birds by using the toxicity centile rank method. $R=1, 2, 3, 4$, and $N=11$. The relevant values are given in Table 6. Based on the values in Table 6 and equations (5–8), the calculated results were $S=10.2$, $L=-0.57$, $A=1.71$, and TRV = RC = 5.5 pg TEQs/g wm.

Four of the lowest TDI values for ring-necked pheasant, American kestrel, screech owl, and chicken were selected to calculate TRG for birds by using the toxicity centile rank method. $R=1, 2, 3, 4$, and $N=10$. The relevant values are presented in Table 7. Based on the values in Table 7 and equations (5–8), the calculated results were $S=9.82$, $L=-1.93$, $A=0.27$, and TDI = 1.31 pg TEQs/kg/day. Using the values of FI:bm for three representative birds (0.23, 0.34, 0.43) in China, the respective RC values were calculated to be 4.53, 3.05, and 5.70 pg TEQs/g wm food. The geometric mean of these three RCs was 4.3 pg TEQs/g wm food, and this value represents the TRG for birds.

5 Results and Discussion

PCBs, which are persistent, bioaccumulative, and toxic are widely distributed in the environment. Apical predators at the top of the aquatic food web, such as fish-eating birds are exposed to greater concentrations of PCBs than are primary and secondary producers. TRGs and TRVs for PCBs in birds derived in this study by SSD, CSA, and TPRM were 10.7, 42.3, 4.3 pg TEQs/g diet wm, and 15.5, 16.7, 5.5 pg TEQs/g tissue wm, respectively (see Table 8). The values derived by the three methods had certain differences, which may have resulted from differences in the toxicity data and calculation methods used. The values of TRGs and TRV derived by using the TPRM were smaller than those determined by applying the other two methods. Because the TPRM used the four lowest toxicity data values for the most sensitive species, the criterion calculated was small and might be over-protective for avian species. However, the values derived from all three methods were similar. The CSA has greater uncertainty because it relies on fewer studies and the uncertainty factor for it is based on judgment and experience. When deriving TRG and TRVs for PCBs in birds by using the SSD approach, PCB toxicity data on about ten avian species were employed. Therefore, the TRG of 10.7 pg TEQs/g diet wm and TRV of 15.5 pg TEQs/g tissue wm, derived by using the SSD method, were recommended as criteria for protecting aquatic birds in China from PCBs.

A TRG of 2.4 pg TEQs/g food wm for PCBs was developed in Canada for protecting avian species that consume aquatic biota (CCME 2001). This TRG was based on a PCB toxicity study in white leghorn chickens, which is one of the most sensitive avian species to PCBs (Barron et al. 1995). As a result of this sensitivity, this TRG would likely be overprotective for wild birds. The TRG value for PCBs in birds derived in this study was 10.7 pg TEQs/g wm food, which is slightly greater than 2.4 pg TEQs/g food wm. Body masses and rates of ingestion of food for three representative avian species were used to derive the PCB TRG values for China. Thus, the TRGs derived by using SSD and TPRM were regarded to be more reasonable than the Canadian TRG for performing risk assessments of PCBs on wild birds in China.

The TRV for effects of PCBs on birds, based on concentrations in tissues (including eggs) developed in this study, was 15.5 pg TEQs/g wm. This value was slightly higher than the TRVs for $TEQ_{WHO\text{-}Avian}$ (0.8–2.9 pg/g wm) that were used to assess ecological risk of great horned owls exposed to PCDD/DF (Coefield et al. 2010).

However, the PCB toxicity data for birds were limited, and uncertainties existed in deriving TRGs and TRVs for PCBs in birds. Food web structure and environmental factors affect the exposure and effects of birds to PCBs. Therefore, further research into the potential for toxic effects of PCBs on birds in China is needed. In addition, more studies of the structure of food webs for avian species in China are needed.

Table 8 The TRGs and TRVs of PCBs for birds by three methods

Methods	SSD	CSA	TPRM
Tissue (pg TEQs/g)	15.5	16.7	5.5
Diet (pg TEQs/g food)	10.7	42.3	4.3

6 Assessment of the Risk PCBs Pose to Birds

6.1 Comparison of TRVs to PCB Concentrations in Birds

As top predators, aquatic birds can accumulate high concentrations of persistent organic compounds, such as PCBs and thus, are often used as receptors of concern in ecological risk assessments. The embryo is the most sensitive life stage for a number of pollutants. Concentrations of pollutants reach young birds primarily via the diet of the female, and PCB concentrations in bird bodies have been found to not correlate with age. PCB concentrations that have been detected in birds from various areas of the world have been summarized in Table 9.

PCB concentrations detected in egrets collected from southern China were approximately 900–3,800 ng/g lm, and the TEQs of PCBs in birds from Hong Kong were greatest (Lam et al. 2008). PCB concentrations in eggs of egrets and black crown night herons from Hong Kong contained levels of 960 (270–1,700) ng/g wm and 230 (85–600) ng/g wm, respectively (Connell et al. 2003).

PCBs in black crown night herons from Chicago contained 586.4–4,678.9 ng/g wm, with an average level of 2,229.6 ng/g wm (Levengood and Schaeffer 2010). The TEQs for the PCBs were not given in these studies. For avian species, PCB concentrations were highest in common tern from the Netherlands and Belgium, followed by cormorants from Japan.

The TEQs for PCBs in birds from Michigan were 1 pg/g wm in eastern bluebirds to 247 pg/g wm in tree swallows, which were comparatively less than those in birds from Japan (17 pg/g wm in whimbrels to 691 pg/g wm in gray herons) (Table 9). TEQs of PCBs in common terns from the Netherlands and Belgium had the greatest level (997 pg/g wm). Based on tissue concentrations, the TRVs of PCBs derived in this study were 5.5–16.7 pg TEQs/g, wm. Most PCB concentrations in birds were

Table 9 Actual PCB concentrations detected in birds from different geographic zones

Zones	Avian species	PCBs (ng/g)	TEQs (pg/g)	Reference
Michigan	House wren	24	10	Fredricks et al. (2010)
	Tree swallow	110	247	Fredricks et al. (2010)
	Eastern bluebird	8	1	Fredricks et al. (2010)
	Great blue heron	223	130	Seston et al. (2010)
Japan	Cormorant	8,327	409	Guruge et al. (2000)
	Gray heron	30	691	Senthilkumar et al. (2002)
	Spot-billed duck	20	37	Senthilkumar et al. (2002)
	Whimbrel	30	17	Senthilkumar et al. (2002)
	Short-tailed shearwater	3	24	Senthilkumar et al. (2002)
	Cattle egret	342	266	Senthilkumar et al. 2002)
	Great egret	504	134	Senthilkumar et al. (2002)
The Netherlands and Belgium	Common tern	43,586	997	Bosveld et al. (1995)

greater than these TRVs. Therefore, it may be that some species of wild birds are experiencing harmful effects from PCB exposure, which is consistent with reported incidents of PCB effects on birds.

6.2 Comparison of TRGs to PCB Concentrations in Fish

PCB concentrations were measured in 20 species of fish (i.e., ten each from fresh and marine waters) from aquatic environments of the Pearl River Delta in China (Wei et al. 2011). Results showed that levels of PCBs in fish ranged from 0.065 to 5.25 pg TEQ/g wm. Based on the levels of PCBs in fish sampled from the Hudson River and New York Bight (Hong and Bush 1990), the TEQs were calculated to be from 0.47 to 6.86 ng TEQs/g wm, which were much higher than those in China.

The TRGs of PCBs calculated in this study were 4.3–42.3 pg TEQs/g food wm, which were higher than most PCB concentrations in fish from the Pearl River Delta in China (0.065–5.25 pg TEQ/g wm). It was indicated that food consumption would not cause harmful effects to birds. But the PCB concentrations in fish from the Hudson River and New York Bight were higher than the TRGs derived in this study, which showed that harmful effects would be caused to birds from food exposure.

7 Summary

PCBs are typical of persistent, bioaccumulative and toxic compounds (PBTs) that are widely distributed in the environment and can biomagnify through aquatic food webs, because of their stability and lipophilic properties. Fish-eating birds are top predators in the aquatic food chain and may suffer adverse effects from exposure to PCB concentrations.

In this review, we address the toxicity of PCBs to birds and have derived tissue residue guidelines (TRGs) and toxic reference values (TRVs) for PCBs for protecting birds in China. In deriving these protective indices, we utilized available data and three approaches, to wit: species sensitivity distribution (SSD), critical study approach (CSA) and toxicity percentile rank method (TPRM). The TRGs and TRVs arrived at by using these methods were 42.3, 10.7, 4.3 pg TEQs/g diet wm and 16.7, 15.5, and 5.5 pg TEQs/g tissue wm for the CSA SSD and TPRM approaches, respectively. These criteria values were analyzed and compared with those derived by others. The following TRG and TRV, derived by SSD, were recommended as avian criteria for protecting avian species in China: 10.7 pg TEQs/g diet wm and 15.5 pg TEQs/g tissue wm, respectively. The hazard of PCBs to birds was assessed by comparing the TRVs and TRGs derived in this study with actual PCB concentrations detected in birds or fish.

The criteria values derived in this study can be used to evaluate the risk of PCBs to birds in China, and to provide indices that are more reasonable for protecting

Chinese avian species. However, several sources of uncertainty exists when deriving TRGs and TRVs for the PCBs in birds, such as lack of adequate toxicity data for birds and need to use uncertainty factors. Clearly, relevant work on PCBs and birds in China are needed in the future. For example, PCB toxicity data for resident avian species in China are needed. In addition, studies are needed on the actual PCB levels in birds and fish in China. Such information is needed to serve as a more firm foundation for future risk assessments.

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