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Chapter 8

Persistent Organic Pollutants in Waterbirds with Special Reference to Hong Kong and Mainland China

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Abstract

Persistent organic pollutants (POPs) have been widely used as pesticides and/or industrial chemicals. Because most POPs are persistent, toxic and bioaccumulative, the United Nations Environment Programme (UNEP) has implemented an approach to reduce/eliminate these substances. A set of 12 POPs have been targeted for global restriction of production/use under Stockholm Convention. The 12 POPs are: aldrin, endrin, dieldrin, dichlorodiphenyltrichoroethanes, polychlorinated biphenyls, polychlorinated dibenzo-*p*-dioxins, polychlorinated dibenzofurans, chlordane, toxaphene, heptachlor, hexachlorobenzene, and mirex. Predatory waterbirds are susceptible to bioaccumulation of POPs through the ingestion of contaminated food sources. They are long-lived and top trophic-level animals in the food web, consequently they are able to integrate pollutant levels over a broad area by bioaccumulation. Thus, they can serve as useful bioindicators for environmental monitoring. This report reviews the relevant literature on the environmental levels and biological effects of the 12 priority POPs, listed under the Stockholm Convention, to waterbirds. Particular attention will be given to the pollution status of POPs in Hong Kong, their threats to waterbirds.

8.1. Background

Hong Kong, located at the mouth of the Pearl River in south China, receives persistent toxic contaminants into marine habitats from a variety of sources including shipping activity, wastewater, industrial wastes and agricultural runoff. Long-term, and often indiscriminate discharges of such contaminants as petroleum hydrocarbons, polycyclic aromatic hydrocarbons, and organochlorine pesticides have compromised water

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and sediment quality, with potentially detrimental implications for environmental integrity, and human and wildlife health. The rapid development of industrial and urban areas in the hinterland of the Pearl River Delta is also an increasingly important source of contaminants to Hong Kong, especially via riverine and aerial inputs. The main purpose of this report is to provide a comprehensive review of the relevant literature to collate and analyze information and data on the environmental levels and biological effects of 12 specific persistent organic pollutants (POPs), listed under the Stockholm Convention, to waterbirds. Particular attention will be given to the pollution status of POPs in Hong Kong, their threats to waterbirds.

Since the Second World War the manufacture and use of organic and inorganic chemicals have increased in both agricultural and industrial areas, and many of these chemicals are released into the environment. There is a considerable concern amongst governments, scientists and nongovernmental organizations over the adverse effects on the environment, wildlife and humans from exposure to chemicals, such as organochlorines. These chemicals are persistent and accumulate along food chains and are often found in natural environments (Barrett et al., 1985). The long life span and the potential for long-range transport of certain chemicals require government intervention to control such chemicals.

It has been widely reported that the decline of bird populations is associated with an increase in the use of organochlorine pesticides, such as DDTs, for insect control in urban and agricultural environments (Blus et al., 1979; Cade et al., 1971; Peakall, 1974). It is also known that some chemicals, in particular organochlorines, interfere with the functioning of reproductive, endocrine, immune, and nervous systems (Yamashita et al., 1993; Jiménez, 1997).

In 1997, the United Nations Environment Programme (UNEP) Governing Council decided that immediate international action should be taken to protect human health and the environment. International negotiations to reduce and eliminate the emission and discharges of an initial set of 12 POPs were initiated at the Stockholm Convention on POPs in May 2001. The 12 substances that were addressed at the Stockholm Convention were: aldrin, endrin, dieldrin, dichlorodiphenyltrichoroethanes (DDTs), polychlorinated biphenyls (PCBs), polychlorinated dibenzo-*p*-dioxins (PCDDs), polychlorinated dibenzofurans (PCDFs), chlordane, toxaphene, heptachlor, hexachlorobenzene (HCB) and mirex. Basic information and status of major organochlorine pesticides in China are summarized in Table 8.1.

Waterbirds are valuable and useful for environmental monitoring. They are particularly susceptible to environmental contaminants because they live long and are top trophic-level animals in the food web.

Table 8.1. Basic information and status of organochlorine pesticides in China

Organochlorine pesticides	CAS no.	Start of use	Ban of use	Total production	Specific exemption	Ban of use in Hong Kong	References
Aldrin	309–00–2	Not used	1983	–	–	1988	Wong et al. (2005) and the references therein
Dieldrin	60–57–1	Not used	1983	–	–	1988	Same as above
Endrin	72–20–8	Not used	–	–	–	Not registered	Same as above
Chlordane	57–74–9	1945	1999	–	Termite control: 95% for structures of houses, 1% for dams and the rest for cable boxes.	1991	
DDT	50–29–3	–	1983	0.27 million tons (1951–1983)	–	1988	Li et al. (2001)
Heptachlor	76–44–8	1948	1982	–	–	Not registered	Wong et al. (2005) and the references therein
HCB	118–74–1	1945	–	–	–	Not registered	Same as above
Mirex	2385–85–5	1958	–	–	20–30% of mirex for termite control in structures of houses, dams and underground cable boxes.	1997	Same as above
Toxaphene	8001–35–2	1948	1982	–	–	1984	Same as above

Consequently, they are able to integrate pollutant levels over a broad area by bioaccumulation (Furness, 1993). A bird egg, unlike a mammalian fetus, is an isolated and independent metabolic system. In addition, persistent, bioaccumulative and lipophilic pollutants (e.g. PCBs, polychlorinated dibenzodioxins and furans) are known to biomagnify in the egg yolk and result in concentrations of orders of magnitude higher than the ambient concentrations in the diet of the female bird (Kleinow et al., 1999). The contaminant levels in waterbird eggs, therefore, provide important and useful information for monitoring changes in environmental quality.

Although there are many peer-reviewed publications about organochlorine pesticides, PCBs and PCDDs/PCDFs, on or in water, sediments and mussels, information about these compounds on or in waterbirds in China is limited. In the present review, most of the data that were presented from China were from limited peer-reviewed publications (Connell et al., 2003; De Luca-Abbott et al., 2001; Dong et al., 2004; Nakata et al., 2005; Wan et al., 2006). Furthermore, it should be noted that the concentrations of various POPs reported were measured in different tissues of separate species, and expressed in different units. Thus, caution should be taken when the information is used or interpreted.

8.1.1. Hexachlorobenzene (HCB)

HCB is an organochlorine product. It was first introduced in 1933 as a fungicide for seed treatment of onion, sorghum and crops, such as wheat, barley, oats and rye, and was used to make fireworks, ammunition and synthetic rubber (Barber et al., 2005; UNEP Chemicals, 2002). It is currently speculated that HCB originates in the environment as a by-product or is the result of impurity in the production of certain chlorinated pesticides, particularly lower chlorinated benzenes and industrial chemicals; the manufacture and application of HCB-contaminated pesticides; and the combustion of waste (Barber et al., 2005; UNEP, 2003; Voldner & Smith, 1989).

The peak of HCB production was between the late 1970s and the early 1980s worldwide (Barber et al., 2005). HCB was introduced in China in 1945, but it was not used as a pesticide (Wong et al., 2005). Rather, it was used as an intermediary in the production of other chlorinated substances, such as sodium-pentachlorophenol (Na-PCP). No information is available on HCB levels in seabirds in China.

Predatory birds, such as cormorants, eagles and guillemots have been used as biomonitoring tools. In the Arctic, the cold annual temperatures and the cold condensation at the higher latitudes may facilitate the deposition and accumulation of HCB (Wania & Mackay, 1993). This hypothesis was further supported by Braune et al. (2001) that the concentration of HCB was higher, for example, in thick-billed murre eggs at higher latitudes. Rocque & Winker (2004) provided evidence for the long-range transport of HCB in cormorants on the Aleutian Islands. The level of HCB declined from west to east in the North Pacific, which suggested that the distribution patterns of HCB might be attributed to industries in Asia, from where it had been transported.

In Antarctica, several studies have reported the concentration of HCB in the eggs of penguins (Corsolini et al., 2002, 2006). A recent study that was conducted by Corsolini and co-workers reported a relatively high concentration of HCB in penguin eggs (Corsolini et al., 2006). However, it was lower than those of the European Arctic and Canadian Arctic seabirds (Table 8.2) (Borgå et al., 2001). The HCB concentration that was found in migratory birds has provided more information about the HCB contamination of their overwintering grounds, though the real situation may be more complex (Muir et al., 1999). Very high concentrations of HCB (up to 10,700 ng g^{-1} dry wt.) were found in birds (e.g. the shoveler) that wintered in China, whereas much lower mean concentrations (10 ng g^{-1} dry wt.) were found in resident birds (e.g. the mallard and the Slavonian grebe). This suggested that significant sources of HCB existed in China (Lebedev et al., 1998). The concentrations of HCB in resident birds collected in the Philippines, India, Vietnam, South India and Japan were compared (Table 8.2) (Kunisue et al., 2003; Minh et al., 2002; Tanabe et al., 1998). Japan had the greatest concentrations of HCB, whereas the Philippines had the lowest concentrations.

In Alaska, HCB concentrations remained relatively stable in Bogoslof and St. George common murre eggs from 1973 to 1976, whereas concentrations of other organochlorines declined (Vander Pol et al., 2004). This phenomenon reflected the continuous production of HCB as a by-product in the manufacture of some industrial chemicals and incineration of waste (Vander Pol et al., 2004). In the Great Lakes, the levels of HCB in the eggs of herring gulls declined significantly by over 90% between 1974 and 2001 (Barber et al., 2005). The reductions of HCB by 70, 90 and 98%, respectively, were observed in the St. Lawrence, Niagara and Detroit Rivers (Barber et al., 2005). The levels of HCB in the bird eggs of black-legged kittiwakes and thick-billed murres in the Canadian Arctic decreased by 64% and 55%, respectively, between 1975 and 1998 (Braune et al., 2001). In the Barents Sea north of Norway, the levels of HCB in the eggs of herring gulls decreased by 37% between 1983 and 1993 (Barrett et al., 1985, 1996). The HCB levels decreased slowly in black guillemots in Iceland

Table 8.2. Hexachlorobenzene (HCB) concentrations in bird samples from different countries

Location	Species	Year	*n*	Tissue	Concentration (ng g^{-1} lipid wt. corrected)	References
Ross Sea, Antarctica	Adelie penguin (*Pygoscelis adeliae*)	1995–1996	6	Egg	197	Corsolini et al. (2006)
Edmonson Point, Antarctica		1996	5	Egg	< 100	Corsolini et al. (2002)
Canadian Arctic	Black-legged kittiwake (*Rissa tridactyla*)	1993–1998	30	Egg	281	Braune et al. (2001)
	Northern fulmur (*Fulmarus glacialis*)	1993–1998	30	Egg	363	
	Thick-billed murre (*Uria lomvia*)	1993–1998	30	Egg	389	
European Arctic	Black guillemot (*Cephyus grylle*)	1995	10	Liver	340	Borgå et al. (2001)
	Brunnich's guillemot (*U. lomvia*)	1995	10	Liver	580	
	Black-legged kittiwake (*R. tridactyla*)	1995	11	Liver	870	
	Glaucous gull (*Larus hyperboreus*)	1995	15	Liver	4100	
Philippines	Painted snipe (*Rostratula benghalensis*)	1994	6	Whole body	1.7	Kunisue et al. (2003)
	Chinese little bittern (*Ixobrychus sinesis*)	1994	3	Whole body	3.0	
	Schrenck's little bittern (*Ixobrychus eurhythmus*)	1994	2	Whole body	4.4	
	Green backed heron (*Butrides striatus*)	1994	3	Whole body	4.9	
India	House crow (*Corvus splendens*)	1995	2	Whole body	4.2	Tanabe et al. (1998)
	Little egret (*Egretta garzetta*)	1995	1	Whole body	10	
	Pond heron (*Ardeola grayii*)	1995	2	Whole body	8.3	
	White-breasted kingfisher (*Halcyon smymensis*)	1995	1	Whole body	3	

	Black-winged stilt (*Himantopus himantopus*)	1995	1	Whole body	13	
	Kentish plover (*Charadrius alexandrinus*)	1995	5	Whole body	8.5	
	Little ringed plover (*Charadris dubius*)	1995	5	Whole body	5.5	
	Kentish plover (*C. alexandrinus*)	1998	2	Whole body	3.3	Kunisue et al. (2003)
Vietnam	Black-capped kingfisher (*Halcyon pileata*)	1997	2	Whole body	26	Minh et al. (2002)
	Common kingfisher (*Alcedo atthis*)	1997	7	Whole body	28	
	White-throated kingfisher (*Halcyon smyrnensis*)	1997	1	Whole body	11	
Vietnam	Common moorhen (*Gallinula chloropus*)	1997	1	Whole body	110	Minh et al. (2002)
	Cinnamon bittern (*Ixobrychus cinnamomeus*)	1997	1	Whole body	25	
	White-breasted waterhen (*Amaurornis phoenicurus*)	1997	3	Whole body	6.9	
Japan						
Shinobazu pond	Common cormorant (*Phalacrocorax carbo*)	1993	8	Liver	4300	Guruge et al. (1997)
Lake Biwa	Common cormorant (*P. carbo*)	1993	9	Liver	360	
Japan	Golden eagle (*Aquila chrysaetos*)	1993–1995	3	Breast muscle	220	Kunisue et al. (2003)
Japan	Golden eagle (*A. chrysaetos*)	1996–1998	8	Liver	1200	
Hokkaido	White-tailed sea-eagle (*Haliaeetus albicilla*)	1997	2	Liver	200	
Hokkaido	White-tailed sea-eagle (*H. albicilla*)	1998	1	Breast muscle	220	

between 1976 and 1996, with a half-time of 16.1 years (Ólafsdóttir et al., 2005).

8.1.2. Heptachlor and chlordane

8.1.2.1. Heptachlor

Heptachlor was isolated from technical chlordane in 1946 and was introduced as an insecticide in the United States in 1952 (USEPA, 1986a; WHO, 1984a). Technical-grade heptachlor contains ~70% heptachlor and 30% related compounds that include 20% chlordane (WHO, 1995). It is a nonsystemic stomach and contact insecticide, which is primarily used against soil insects and termites (WHO, 1984a). It has also been used against cotton insects, grasshoppers, crop pests and to combat malaria (Smith, 1991; WHO, 1995). Heptachlor epoxide is an oxidation product of heptachlor and chlordane (WHO, 1995). The toxicities of heptachlor and heptachlor epoxide are similar and they bioaccumulate in the fat of organisms.

It is difficult to determine the actual amount of heptachlor that is used; however, the total use of heptachlor in the United States was estimated to be 16,000 tons between 1971 and 1976 (Fendick et al., 1990). Due to the adverse effects of heptachlor, most commercial, agricultural and domestic uses of heptachlor in the United States were phased out between 1974 and 1988, and all uses of heptachlor products were banned by the United States Environmental Protection Agency (USEPA) in 1988 (USEPA, 1990a). The only current use of heptachlor is in the treatment of fire ants in power plants (ATSDR, 2005). Heptachlor was used in 1948 in China and it was banned in 1982 (Wong et al., 2005).

In the 1970s, heptachlor was extensively used, and caused serious problems in wild birds. The applications of granular heptachlor in Georgia and heptachlor in Mississippi caused the long-term decline in the population of northern bobwhite quail (*Colinus virinanus*) and a short-term abrupt decline in the population of several passerines (Ferguson, 1964; Rosene, 1965). Several wild bird populations, including Canada geese and American kestrels in the Columbia Basin in the United States, declined due to heptachlor (Blus et al., 1984; Henny et al., 1983). Heptachlor epoxide residues that were found in the brains of dead Canada geese at the Umatilla National Wildlife Refuge in Oregon in 1978 and 1979 were equal to or exceeded the lethal hazard zone level (8–9 $\mu g\ g^{-1}$ wet wt.). Moreover, the residues that were found in eggs were greater than 10 $\mu g\, g^{-1}$ wet wt., which was related to low nest success (Blus et al., 1984). The source of exposure was thought to be heptachlor treated seeds that were consumed by the geese. After a substitution of lindane with heptachlor, the reproductive success of geese increased and adult mortality decreased. The nesting population increased from 102 pairs in 1979 to 170 pairs in 1983 (Blus et al., 1984). The reproductive success of American kestrels in the same area decreased (Henny et al., 1983). Heptachlor epoxide residues that were found in the eggs were greater than $1.5\,\mu g\ g^{-1}$ wet wt. and such concentrations were associated with the reduced productivity. As kestrels were not seed eaters, heptachlor epoxide bioaccumulates along the food chain through eating geese (Blus et al., 1984). Dong et al. (2004) reported that the heptachlor and heptachlor epoxide levels of heron eggs from Lake Tai in China were up to $126\,ng\ g^{-1}$ dry wt. but the occurrence of these compounds was low.

8.1.2.2. Chlordane

Chlordane is a mixture of at least 120 compounds and technical chlordane typically contains 64–67% chlorine (Dearth & Hites, 1991a,b; WHO, 1995). Of these 120 compounds, 60–75% are chlordane isomers (*cis*- and *trans*-) and the remainder is related to endo-compounds that include heptachlor, nonachlor, diels-alder adduct of cyclopentadiene and penta/hexa/octachlorocyclopentadienes (UNEP Chemicals, 2002). Heptachlor contributes up to 10% of technical chlordane and it is also a pesticide formulation.

Chlordane was introduced as an insecticide in 1945 and was the first cyclodiene insecticide that was used in agriculture (Eisler, 1990). It was the second most important organochlorine pesticide after toxaphene from 1976 to 1977 (Stansley & Roscoe, 1999). It has been used on agricultural crops and extensively in the control of termites (Smith, 1991). Chlordane and heptachlor can be metabolized into two persistent (oxygenated) epoxides—oxychlordane and heptachlor epoxide—in mammals (Nomeir & Hajjar, 1987) such that the two compounds are always measured together with chlordane and heptachlor.

The production of chlordane was reduced from ~3.5 to 4.0 million pounds in 1986 to 100,000 to 1 million pounds in 1991 (ATSDR, 1994a). More than 63 million kg of chlordane were produced and used in the United States, mostly after 1960 before sales and its use were suspended in 1988 (Dearth & Hites, 1991a,b). Restrictions were imposed on the use of chlordane in 1979 because of its potential human carcinogenicity. After this time chlordane was used mainly for underground termite control and in building construction (Dearth & Hites, 1991a,b). In Japan, chlordane was only permitted for the control of termites and powder post beetles (Miyazaki et al., 1980). USEPA cancelled its registration for commercial production, delivery, sale, and use in 1988 (USEPA, 1988). The use of chlordane was introduced in China in 1945 but it was banned in 1999 (Wong et al., 2005). With special exemption, it is still produced in China and is used locally for termite control—95% for structures of houses, 1% for dams and the remainder for cable boxes.

The first chlordane-related mortality was of three wild birds and was recorded between 1978 and 1981 (Blus et al., 1983). The levels of chlordanes and heptachlor epoxide from the two adult male red-shouldered hawks (*Buteo lineatus*) and an adult female great horned owl (*Bubo virginianus*) were within the critical lethal range that has been defined by experimental studies (heptachlor epoxide in brain tissue: 3.4–8.3 $\mu g\ g^{-1}$ wet wt.; oxychlordane in brain tissue: 1.1–5.0 $\mu g\ g^{-1}$ wet wt.). The chlordane poisoning of birds has been reported in several studies in the United States (Blus et al., 1983, 1985; Post, 1951; Stansley & Roscoe, 1999). From 1986 to 1990, 122 cases of avian mortality due to chlordane and/or dieldrin were documented in New York, Maryland and New Jersey (Okoniewski & Novesky, 1993). High pesticide concentrations were found in cyclodiene-resistant insect populations. These pesticide-tainted insects, when eaten by birds, caused mortalities in the avian populations (Okoniewski & Novesky, 1993).

Comparisons of data are difficult as different studies have reported total chlordanes with different compositions (e.g. sum of oxychlordane, heptachlor epoxide, *trans*-chlordane, *cis*-chlordane, *trans*-nonachlor, *cis*-nonachlor, MC5 and MC7 vs. sum of oxychlordane, heptachlor epoxide, *trans*-nonachlor) and with different tissue samples. Attention should be paid to the definition of total chlordanes because it has different meanings. In the following paragraphs, the concentrations of the sum of chlordane and its metabolites, heptachlor and heptachlor epoxide, are both discussed. Oxychlordane, *trans*-nonachlor and *cis*-nonachlor contributed to the major components of total chlordane in bird samples (Table 8.3) (Fisk et al., 2001; Stansley & Roscoe, 1999). The levels of oxychlordane in the livers and eggs of the bird samples that were collected in the Canadian high Arctic between 1975 and 1976 were similar and lower, respectively, than those of the samples that were collected in the high Arctic of Northwater Polynya, in 1998 (Fisk et al., 2001). Another study showed that there was a significant decrease of total chlordanes (the sum of oxychlordane, heptachlor epoxide, *trans*-chlordane, *cis*-chlordane, *trans*-nonachlor, and *cis*-nonachlor) in the eggs of black-legged kittiwakes from 1975 to 1998 (Braune et al., 2001). Total chlordanes (the sum of oxychlordane, trans-nonachlor and heptachlor epoxide) that were found in the livers of glaucous gulls that were collected in the southern Barents Sea in 1991 were similar to those that were found in

Table 8.3. Total chlordane concentrations in bird samples from different countries

	Location	Species	Year	*n*	Tissue	Concentration (ng g^{-1} lipid wt. corrected)	References
1975–1998	Canadian Arctic	Black-legged kittiwake (*Rissa tridactyla*)	1975	12	Egg	490	Braune et al. (2001)
			1976	6	Egg	1400	
			1993	15	Egg	390	
			1998	15	Egg	360	
1990s–now	Southern Barents Sea, Arctic	Black-legged kittiwake (*R. tridactyla*)	1991	19	Liver	780	Savinova et al. (1995)
		Black-legged kittiwake (*R. tridactyla*)	1998	10	Liver	660	Fisk et al. (2001)
	Northwater Polynya, Arctic	Glaucous gull (*Larus hyperboreus*)	1991	10	Liver	2300	Savinova et al. (1995)
	Southern Barents Sea, Arctic	Glaucous gull (*L. hyperboreus*)	1998	11	Liver	2200	Fisk et al. (2001)
	Ross Sea, Antarctica	Adelie penguin (*Pygoscelis adeliae*)	1995–1996	6	Egg	19	Corsolini et al. (2006)
	Canadian Arctic	Black-legged kittiwake (*R. tridactyla*)	1993–1998	30	Egg	370	Braune et al. (2001)
		Northern fulmur (*Fulmarus glacialis*)	1993–1998	30	Egg	660	
		Thick-billed murre (*Uria lomvia*)	1993–1998	30	Egg	220	
	Philippines	Green backed heron (*Butrides striatus*)	1994	3	Whole body	78 (44–110)	Kunisue et al. (2003)
	India	Long-billed Mongolian plover (*Chradrius mongolus*)	1995	6	Whole body	170	Tanabe et al. (1998)

Table 8.3. (*Continued*)

Location	Species	Year	*n*	Tissue	Concentration (ng g^{-1} lipid wt. corrected)	References
India	Little tern (*Sterna albifrons*)	1995	2	Whole body	27	Tanabe et al. (1998)
	Short-billed Mongolian plover (*Chradrius mongolus*)	1995	6	Whole body	29	
Lake Baikal, Russia	Grey heron (*Ardea cinerea*)	1996	2	Breast muscle	27	Kunisue et al. (2002)
Japan						
Shinobazu pond	Common cormorant (*Phalacrocorax carbo*)	1993	8	Liver	9600	Guruge et al. (1997)
Lake Biwa	Common cormorant (*P. carbo*)	1993	9	Liver	1300	
Japan	Golden eagle (*Aquila chrysaetos*)	1993–1995	3	Breast muscle	3300	Kunisue et al. (2003)
	Golden eagle (*A. chrysaetos*)	1996–1998	8	Liver	14,000	
Hokkaido	White-tailed sea-eagle (*Haliaeetus albicilla*)	1997	2	Liver	2700	
	White-tailed sea-eagle (*H. albicilla*)	1998	1	Breast muscle	3000	
Hong Kong	Little egret (*Egretta garetta*)	2000	9	Egg	4300 (1230–7100)	Connell et al. (2003)
	Black-crowned night heron (*Nycticorax nycticorax*)	2000	9	Egg	460 (77–1300)	

Northwater Polynya in 1998 (Fisk et al., 2001; Savinova et al., 1995). For the penguin eggs that were collected in Antarctica between 1995 and 1996, total chlordanes (sum of oxychlordane, *trans*-chlordane, *cis*-chlordane, and *trans*-nonachlor) were much lower than those of the bird eggs from the Arctic (Braune et al., 2001; Corsolini et al., 2006). The levels of chlordanes in resident bird tissue samples from the Philippines, India, Russia and Vietnam were similar, whereas those of Japan and Hong Kong had high levels of chlordanes (Table 8.3) (Connell et al., 2003; Guruge et al., 1997; Kunisue et al., 2002; Minh et al., 2002; Tanabe et al., 1998). In conclusion, Japan and Hong Kong had the highest concentrations and were followed by the Artic region, which was higher than that of the Philippines, India, Vietnam and Russia, while the Antarctic region had the lowest concentration.

8.1.3. Mirex

Mirex was first synthesized in 1946 but it was not introduced as a pesticide against hymenopterous insects, especially ants, until 1959 (Smith, 1991). Technical grade mirex consists of approximately 95% mirex and less than 2.5% chlordecone, mostly kepone (Eisler, 1985). Mirex is a stomach insecticide with little contact activity. The main use of mirex was against the imported fire ants in the south-eastern United States (WHO, 1984b). It has also been used to control leaf cutters in South America, harvester termites in South Africa, Western harvester ants in the United States, the pineapple mealy bug in Hawaii and it was proposed to have been used against yellow jacket wasps in the United States (WHO, 1995). Under the trade name of Dechlorane, mirex was used as a fire retardant in electronic components, fabrics, rubber, plastics and electrical goods (Eisler, 1985; WHO, 1995).

The total sales of mirex by Hooker Chemicals and Plastics Corporation, in the United States between 1959 and 1975, were around 1,528,000 kg (Kaiser, 1978). Among the total sales of mirex, 26% was for insecticidal use and 76% was for other uses, such as the incorporation of mirex into plastics to improve flame- and fire-retardant properties (Kaiser, 1978). Between 1961 and 1975, ~400,000 kg of mirex were used as pesticides, of which approximately 250,000 kg were sold in the south eastern United States for the control of native and imported fire ants (*Solenopsis* spp.) and most of the remainder was exported to Brazil for use in fire ant control (Eisler, 1985).

The concern about mirex became of international importance after its discovery in fish from Lake Ontario (Kaiser, 1978). The mirex contamination in fish from the Bay of Quinte, approximately 50 km upstream from the mouth of this ''tributary'' to Lake Ontario and the nearest producer and processor of the compound, Hooker Chemicals and Plastics Corporation, was located in New York State on the Niagara River, approximately 250 km from the site of the observed fish contamination. This raised the question of whether these findings were due to local contamination or of widespread occurrence (Kaiser, 1978). A herring gull egg was destroyed after pipping the eggshell, and deformation of the beak of a Lake Ontario bird's chick due to mirex was observed (Kaiser, 1978). Photomirex (8-monhydromirex) was formed during degradation and was found in herring gull eggs (Hallett et al., 1976). This derivative has similar chemical and physical properties, and also similar biological potency to mirex (Hallett et al., 1976). In 1978, the United States Environmental Protection Agency banned all uses of mirex as a pesticide (USEPA, 2001). Mirex has been used in China since 1958 and it is still produced and used locally for termite control (Wong et al., 2005).

Limited data are available on mirex worldwide except in the United States. The levels of mirex found in birds that were collected in the United States, except of those from the southeastern United States and the Great Lakes, were low and were considered nonhazardous in the 1970s (Cain & Bunck, 1983). White (1979) investigated the wings of mallards and American black ducks (*Anas rubripes*) that were collected from four major flight pathways between 1976 and 1977 (Eisler, 1985 and the references therein). The results showed that Atlantic mallards had the highest detection frequencies of occurrence at 50% and the highest concentration of mirex (0.14 $\mu g\ g^{-1}$ wet wt.). They were followed by Mississippi mallards at 29% and 0.03 $\mu g\ g^{-1}$ wet wt., Central mallards at 14% and 0.06 $\mu g\ g^{-1}$ wet wt., and Pacific mallards at 4% and 0.03 $\mu g\ g^{-1}$ wet wt. (Eisler, 1985).

Between 1972 and 1973, Ohlendorf and co-workers measured the levels of mirex and other organochlorine compounds in eggs of anhingas (*Anhinga anhinga*), herons, egrets, bitterns, ibises and storks in various locations throughout the eastern United States (Eisler, 1985). The highest mean mirex concentration of 0.74 $\mu g\ g^{-1}$ wet wt. that ranged from 0.19 to 2.5 $\mu g\ g^{-1}$ wet wt., was found in the eggs of green herons (*Butorides striatus*) from the Savannah National Wildlife Refuge in South Carolina and in a single egg of the cattle egret (*Bubulcus ibis*) with a mirex level of 2.9 $\mu g\ g^{-1}$ wet wt. However, the Great Lakes region had the highest overall frequency of mirex occurrence (24%) in eggs among the study areas: the south Atlantic coastal region (15.6%), inland areas (10.7%), the Gulf Coast (4.4%) and the North Atlantic region (3.2%) (Eisler, 1985).

Mirex residues were detected in migratory birds that were collected from various locations and included areas that were far from known sources or applications of mirex (Eisler, 1985). For example, 22% of the eggs from 19 species of Alaskan seabirds that were collected between 1973 and 1976 contained mirex. The highest concentration was 0.044 μg g^{-1} wet wt. in eggs of a fork-tailed storm petrel (*Oceanodroma furcata*) from the Barren Islands. Mirex residues were low compared with those of other organochlorine compounds. The eggs from clapper rails (*Rallus longirostris*) that were collected in New Jersey from 1972 to 1974 contained 0.16 to 0.45 μg g^{-1} wet wt. of mirex (Klaas et al., 1980). The eggs of greater black-backed gulls (*Larus marinus*) that were collected from Appledore Island, in Maine, in 1977 contained up to 0.26 μg g^{-1} wet wt., but no mirex was detected in the eggs of common eiders (*Somateria mollissima*) or herring gulls from the same area. The greater black-backed gull is an active carnivore and the higher mirex levels in black-backed gulls might be attributed to its predatory feeding habits on small birds and mammals (Szaro et al., 1979). The eggs of pigeon hawks (*Falco columbarius*) and peregrine falcons (*Falco peregrinus*) had very high concentrations of mirex (0.25 μg g^{-1} wet wt. and 0.43 μg g^{-1} wet wt., respectively) in the higher latitudes of the Arctic (eastern and northern Canada). These two species feed on migratory birds or may migrate to mirex-affected areas (Kaiser, 1978).

Mirex was also found in the eggs of cormorants (*Phalacrocorax* sp.) from the Bay of Fundy on the Atlantic coast of Canada. The residue levels from 1975 (0.113 μg g^{-1} wet wt.) were double that of earlier levels from 1973 (0.058 μg g^{-1} wet wt.) and 1974 (0.059 μg g^{-1} wet wt.) (Kaiser, 1978). The source of contamination was suspected to be the southern wintering range in Florida and the Gulf of Mexico (Kaiser, 1978).

Few studies were available to evaluate the temporal change of mirex in bird samples. The only studies of mirex in seabird eggs were conducted by Braune et al. (2001) on the temporal trends from 1975 to 1998 in the Canadian Arctic. A decreased trend of mirex levels was only observed in eggs of black-legged kittiwakes but not in northern fulmars and thick-billed murres. The levels of mirex that were measured in the eggs of the studied animals ranged from 0.003 to 0.013 μg g^{-1} wet wt. in 1998. There did not appear to be any consistent change in the proportions over the study period (Braune et al., 2001). No information is available on the level of mirex in waterbirds in China.

Bird et al. (1983) found that when captive American kestrels (*Falco sparverius*) were fed 8 mg kg^{-1} of mirex for 69 days, there was a decline in sperm concentration and a slight increase in semen volume. An overall net decrease of 70% in sperm number was observed. The investigators believed that lower semen quality in the breeding season, coupled with altered courtship, could reduce the fertility of eggs and the reproductive fitness of individuals (Bird et al., 1983). Studies showed that eggshells of mallards that were fed 100 mg kg^{-1} of mirex, thinned and duckling survival was reduced (Waters et al., 1977).

8.1.4. Aldrin and dieldrin

Dieldrin and aldrin were first synthesized in 1948 and they were commercially manufactured as pesticides in 1950 (WHO, 1989). Technical grade aldrin contains 90% 1,2,3,4,10,10-hexachloro-1,4,4α,5,8,8α-hexahydro-1,4-endo,exo-5,8-dimethanonaphthalene (HHDN). Technical-grade dieldrin contains 85% 1,2,3,4,10,10-hexachloro-6,7-epoxy-1,4,4α,5,6,7,8,8α-octahydro-1,4-endo,exo-5,8-dimethanonaphthalene (HEOD). Dieldrin can be synthesized from aldrin (WHO, 1989). As aldrin is readily metabolized into dieldrin by both plants and animals, aldrin residues are only found in trace amounts in plants and animals, if at all (WHO, 1989).

The peak period for the use of aldrin and dieldrin was between the late 1960s and the early 1970s throughout various parts of the world (WHO, 1989). The use pattern of aldrin and dieldrin are quite similar. They act as effective contact and stomach poisons for insects. They are used to control soil insects (e.g. grasshoppers and corn rootworm), and protect crops and wooden structures from termites (WHO, 1989). The production of aldrin and dieldrin has decreased since the early 1960s. In the United States, the peak use of aldrin from 19 million pounds in 1966 decreased to 10.5 million pounds in 1970 (USEPA, 1980). During this same period (1966–1970), annual dieldrin use dropped from 1 million to 670,000 pounds. These decreases were primarily due to increased insect resistance to the aldrin and dieldrin, and to the development and availability of more effective and environmentally friendly pesticides (USEPA, 1980).

The global production of aldrin and dieldrin decreased from 20,000 tons in 1971 to less than 2500 tons in 1984 (WHO, 1989). In 1972, USEPA discontinued all but three specific uses of these compounds for subsurface termite control, ‘‘dipping of nonfood plant roots and tops, and moth-proofing in manufacturing processes in closed systems’’ (USEPA, 2003). In China, aldrin and dieldrin were produced on a small scale for research purposes and were regarded as ‘‘not used’’ (Wong et al., 2005). They were banned in China in 1983.

Between 1963 and 1975, HEOD (dieldrin) accounted for ~50% and 39% of all recorded sparrow hawk and kestrel deaths, respectively, in Britain (Newton et al., 1992). There was a recovery of the populations in between 1987 and 1990 after a marked reduction in aldrin–dieldrin use (Newton et al., 1992). The decline of the bird populations was due to the fact that these chemicals were highly toxic and poisoned birds immediately, which increased the mortality above the natural level (Newton et al., 1992). During the period between 1967 and 1971, a total of 192 bird casualties were recorded along the Texas Gulf Coast. The main cause of the deaths was intoxication due to the intake of aldrin treated rice (WHO, 1989).

Several studies of aldrin and dieldrin on the effect of normal fertility and hatchability on chickens and pheasants have been conducted (WHO, 1995). Normal fertility and hatchability slightly decreased when the chickens were fed a 10 mg dieldrin kg^{-1} diet, whereas when pheasants were fed a 50 mg dieldrin kg^{-1} diet, it caused a significant effect on fertility and hatchability (WHO, 1995). Based on these findings, the WHO (1989) concluded that reproductive success was not consistently affected in the absence of maternal toxicity.

There are declining trends of dieldrin that have been measured from bird samples. Residue levels of dieldrin in unhatched bald eagle eggs that were collected from 1974 to 1980 along Lake Erie, decreased from 1.28 to 0.49 $\mu g\ g^{-1}$ wet wt. as compared with those that were collected from 1989 to 1994 (ATSDR, 2002a). A similar decreasing trend was also observed of the eggs of murres at Bogoslof Island and St. George Island in Alaska between 1973 and 2000 (Table 8.4a) (Vander Pol et al., 2004). In another study of the Canadian Arctic, Braune's group found that there was a decreasing trend of dieldrin in black-legged kittiwakes but there was no decreasing trend in northern fulmurs and thick-billed murres in the same area (Braune et al., 2001).

The levels of dieldrin and aldrin are also studied in the United States, the Canadian Arctic, Greece and Alaska (Table 8.4a). The bird samples from New Jersey from 1996 to 1997 had the highest levels of dieldrin and aldrin (Stansley & Roscoe, 1999), whereas the Canadian Arctic had the lowest levels of dieldrin and aldrin (Braune et al., 2001). A recent study from Greece reported that the concentrations of dieldrin and aldrin from the liver samples of birds were much lower than that of the United States but were similar to those of the Arctic (Sakellarides et al., 2006). Dong et al. (2004) showed that the occurrence of aldrin and dieldrin in herons in China were low as compared with Italy (50–73%) (Table 8.4b) (Fasola et al., 1998) and dieldrin was absent in little egrets and cattle egrets. The authors suggested that aldrin and dieldrin are not used as frequently in China as in Europe.

8.1.5. Endrin

Endrin was introduced in the United States as an insecticide, rodenticide and avicide in 1951 (ATSDR, 1996a). It was also used as an

Table 8.4a. Dieldrin concentrations in bird samples from different countries

Location	Species	Year	*n*	Tissue	Concentration (ng g^{-1} wet wt.)	References
Bogoslof, Alaska	Murre (*Uria aalge*)	1973–1976	7	Egg	34	Vander Pol et al. (2004) and the references therein
St George, Alaska	Murre (*U. aalge*)	2000	9	Egg	2.3	Vander Pol et al. (2004)
Bogoslof, Alaska	Murre (*U. aalge*)	1973–1976	11	Egg	9	Vander Pol et al. (2004) and the references therein
St George, Alaska	Murre (*U. aalge*)	1999	11	Egg	4.2	Vander Pol et al. (2004)
Canadian Arctic	Black-legged kittiwake (*Rissa tridactyla*)	1975	12	Egg	14	Braune et al. (2001)
		1998	15	Egg	9	
	Northern fulmur (*Fulmarus glacialis*)	1975	15	Egg	13	
		1998	15	Egg	14	
	Thick-billed murre (*Uria lomvia*)	1975	9	Egg	14	
		1998	15	Egg	15	
New Jersey, USA	Red tailed hawk (*Buteo jamaicensis*)	1997	2	Brain	1600–3100	Stansley and Roscoe (1999)

	Cooper's hawk (*Accipiter cooperi*)	1996–1997	9	Brain	Up to 2600	
Greece	White stork (*Ciconia ciconia*)	2003	6	Liver	0.086	Sakellarides et al. (2006)
	Greater flamingo (*Phoenicopterus ruber*)	2003	2	Liver	3.9	
	Dalmatian pelican (*Pelecanus crispus*)	2003	1	Liver	28	
	Little bittern (*Ixobrychus minutus*)	2003	10	Liver	1.2	
	Bittern (*Botaurus stellaris*)	2003	1	Liver	7.1	
	Little egret (*Egretta garetta*)	2003	5	Liver	1.1	
Lake Tai, China	Black-crowned night heron (*Nycticorax nycticorax*)	2000	65	Egg	1.13	Dong et al. (2004)
	Little egret (*E. garetta*)	2000	36	Egg	n.d.[a]	
	Cattle egret (*Bubulus ibis*)	2000	13	Egg	n.d.	
	Chinese pond heron (*Ardeola bacchus*)	2000	17	Egg	0.072	

[a]n.d.: Not detected.

Table 8.4b. Percentage occurrence of aldrin, dieldrin and endrin in bird samples

Location	Species	Year	*n*	Tissue	Occurrence (%)			References
					Aldrin	Dieldrin	Endrin	
Lake Tai, China	Black-crowned night heron (*Nycticorax nyticorax*)	2000	65	Egg	4.6	22	9.2	Dong et al. (2004)
	Little egret (*Egretta garetta*)	2000	36	Egg	47	0	0	
	Cattle egret (*Bubulus ibis*)	2000	13	Egg	0	0	7.7	
	Chinese pond heron (*Ardeola bacchus*)	2000	17	Egg	24	35	12	
Northern Italy	Little egret (*E. garetta*)	1993–1994	22	Egg	73	50	27	Fasola et al. (1998)
	Black-crowned night heron (*N. nyticorax*)	1993–1994	7	Egg	100	100	29	
		1982–1983	20	Egg	55	15	30	

insecticide agent on bird perches. Although the chemical properties of endrin (*endo*, *endo* stereoisomer of dieldrin) are very similar to those of aldrin or dieldrin, it had never been used extensively for termite or other applications in urban areas (Blus et al., 1989). Endrin had been one of the major chemicals used for controlling voles (*Microtus* spp.) in orchards (Petrella et al., 1975). Endrin aldehyde and endrin ketone are the impurities or degradation products of endrin, and trace amounts of impurities, such as aldrin and dieldrin, could also be found (ATSDR, 1996a; USEPA, 1985).

Because of the environmental concern and the development of resistance in certain pests, the use of endrin was greatly reduced in the United States and elsewhere (USEPA, 1985). The total sale of endrin in the United States was estimated to be 2.3 million kg in 1962, whereas the total production of endrin was decreased to 450,000 kg in the United States in 1971 (ATSDR, 1996a). The extreme toxicity of endrin affected the nontarget populations of raptors, such as California quail and chukars, and migratory birds in Washington State fruit orchards, and was a major reason for its discontinuation as a pesticide agent (ATSDR, 1996a; Blus et al., 1989). All uses of endrin in the United States were voluntarily discontinued by the manufacturer in 1986, and its use as a toxicant on bird perches was also discontinued in 1991 (ATSDR, 1996a; USEPA, 1983).

There was a disastrous die-off of the transplanted pelicans in Louisiana in 1975 (Blus et al., 1979). Endrin was the major factor for the die-offs, as the residues in the brains of several pelicans were similar to those that were found in the brains of experimental birds that were dying from doses of endrin (Blus et al., 1989). Stickel and co-workers reported that all of the experimental passerines died between 2 and 9 days after they were fed with $10\,\mu g\ g^{-1}$ wet wt. of endrin and the endrin residues in the brains were found to be lethal ($\geqslant 0.8\,\mu g\ g^{-1}$ wet wt.) or in the danger zone ($0.6–0.79\,\mu g\ g^{-1}$ wet wt.) of experimental birds. Fat and pectoral musculature was greatly reduced or absent in all birds or in some, respectively. Between 1979 and 1983, among the 194 birds that were found dead in or near Central Washington Orchards, 6 and 46 of them had $0.6–0.79\,\mu g\ g^{-1}$ wet wt. (danger zone) and greater than $0.8\,\mu g\ g^{-1}$ wet wt. (lethal) endrin residues, respectively, in the brains of the birds (Blus et al., 1989). The applications of endrin in the orchard garden attributed to these casualties. It was also suggested that endrin might have induced serious physiological problems and caused the mortalities from other factors, such as the effect of vehicles or other objects due to effects on vision, predation or disease. However, there was no evidence to demonstrate that endrin depressed reproductive success.

There are limited data available to understand the geographical distribution of endrin residues in bird. Goutner et al. (2001) reported that the levels of endrin residues in the eggs of Audouin's gulls in the north-eastern Mediterranean between 1997 and 1998 was much lower than those of the birds of Blus et al. (1989) between 1981 and 1983 (Table 8.55). In a more recent study that measured the level of endrin residues in the livers and muscles of birds in Greenland, however, the total drin (sum of dieldrin, aldrin and endrin) was given and it was difficult to obtain the level of endrin residues from Greenland biota (Vorkamp et al., 2004). Dong et al. (2004) measured the levels of endrin in herons from Lake Tai in China. The occurrence and levels of endrin in heron eggs were low compared with those of Italy (Fasola et al., 1998).

8.1.6. Toxaphene

Toxaphene has been in use since 1949. It is a complex mixture that consists of at least 670 chlorinated terpenes (Jansson & Wideqvist, 1983). It was used as a nonsystemic stomach and contact insecticide. As it is nontoxic to plants (except to cucurbius), it was used to control many insects that inhabited cotton, corn, fruit, vegetables and small grains, and to control the *Cussia obtusifola* soybean pest. Toxaphene solutions were usually mixed with other pesticides because it can help to solubilise other insecticides with lower water solubility (e.g. DDT, lindane, etc.) (ATSDR, 1996b).

During the early 1970s, toxaphene or mixtures of toxaphene with rotenone were applied in lakes and streams by game fish agencies as a piscicide to remove undesirable fish for sport fishing, however, it was found to cause damage to nontarget organisms (ATSDR, 1996b). The peak consumption of toxaphene occurred in 1972 after DDT was banned. The use of toxaphene declined in the United States drastically after 1975 and it was reported to be the most heavily used pesticide (ATSDR, 1996b). In 1974, $\sim$20 million kg of toxaphene was used in the United States, whereas in 1980, the total use of toxaphene was estimated at around 8.5 million kg and 4.9 million kg in 1982 (WHO, 1984c). In 1990, all registered uses of toxaphene and its mixtures were discontinued in the United States (USEPA, 1990b). China began to use toxaphene in 1958 and the total production of toxaphene from 1964 to 1980 was estimated to be 24,000 tons (Wong et al., 2005). There is no current use and stockpiling of toxaphene after it was banned in 1987.

Generally, the levels of toxaphene and the frequency of occurrence in birds are low or nondetected as compared with other organochlorine

Table 8.5. Endrin concentrations in bird samples from different countries

Location	Species	Year	*n*	Tissue	Concentration (ng g^{-1} wet wt.)	References
Lake Tai, China	Black-crowned night heron (*Nycticorax nyticorax*)	2000	65	Egg	0.42	Dong et al. (2004)
	Little egret (*Egretta garetta*)	2000	36	Egg	n.d.[a]	
	Cattle egret (*Bubulus ibis*)	2000	13	Egg	0.075	
	Chinese pond heron (*Ardeola bacchus*)	2000	17	Egg	0.084	
Northern Italy	Little egret (*E. garetta*)	1993–1994	22	Egg	13	Fasola et al. (1998)
	Black-crowned night heron (*N. nyticorax*)	1993–1994	7	Egg	10	
	Black-crowned night heron (*N. nyticorax*)	1982–1983	20	Egg	110	
North-Eastern Mediterranean	Audouin's gull (*Larus audouinni*)	1997–1998	59	Egg	0.73 (up to 3.3)	Goutner et al. (2001)
Washington, USA	California quail (*Callipepla californica*)	1981–1982	17	Liver	220 (n.d.–5300)	Blus et al. (1989)
	Mallard (*Anas platyrhynchos*)	1981–1982	10	Liver	n.d.–20	
	Canada geese (*Branta canadensis*)	1982	3	Liver	250 (190–380)	

[a]n.d.: Not detected.

pesticides and those in fish (Eisler & Jacknow, 1985). However, there are some extensive reports on toxaphene occurrence. Fifty-five male wild turkeys (*Meleagris gallopavo*) that were collected during the hunting season in southern Illinois in 1974 had up to 0.9 μg g^{-1} wet wt. toxaphene (Eisler & Jacknow, 1985). Toxaphene poisoning was reported from the deaths of two birds in California (Pollock & Kilgore, 1978). The causes of the deaths were due to the results of the biomagnification of toxaphene through food consumption and the direct exposure of toxaphene during a spray for the control of grasshoppers. Bird carcasses contained 0.1–9.6 μg of toxaphene g^{-1} wet wt. 2–3 weeks after the spray. Fetotoxic effects were reported on ring-necked pheasants (Pollock & Kilgore, 1978). Reduced hatchability in eggs was recorded in this bird after it was fed 100 mg toxaphene kg^{-1}. Reduced growth and backbone impairment were observed when ducklings were fed 10 or 50 mg toxaphene kg^{-1} for 90 days (Mehrle et al., 1979).

There is limited information of the levels of toxaphene on avian species, particularly seabirds. From a study in the Baltic region, the level of toxaphenes of the breast muscles of guillemots from 1970 to 1981 did not show any temporal trend (Andersson et al., 1988). The level of toxaphene in another study from the same sampling areas showed that the levels of toxaphene in eggs of guillemot had decreased from 1976 to 1989 (Table 8.6) (Wideqvist et al., 1993). The authors suggested that the negative results of the previous study for a temporal trend might be due to the large variation in the accumulation of contaminants from different sex and age (Wideqvist et al., 1993).

With limited information, generally, the levels of toxaphene were lower in the Polar Regions as compared with those in the north Pacific (Table 8.6). The eggs of skuas (*Catharacta* sp.) and penguins (*Pygoscelis* sp.) that were collected from Antarctica between 1993 and 1994, and the livers of glaucous gulls that were collected from Bear Island and the Arctic of Norway between 1995 and 1999 all contained low levels of toxaphene (Herzke et al., 2003; Muir et al., 2002). In contrast, the eggs of blackfooted albatross that were collected between 1994 and 1995 contained higher toxaphene levels (Muir et al., 2002). Witte et al. (2000) reported a decreasing trend of toxaphene in eggs of common terns in Trischen, northern Germany between 1981 and 1997. In another study, although reduced levels of toxaphenes were observed in the livers of glaucous gulls from Bear Island, no significant decreasing trend was observed (Herzke et al., 2003). Limited materials, large differences in age, different migratory patterns and metabolic processes or choice of food items contributed to the large variation in toxaphene levels, which could have masked any decreasing trends. There is no information available for toxaphene on waterbirds in China.

Table 8.6. Toxaphene concentrations in bird samples from different countries

	Location	Species	Year	*n*	Tissue	Concentration (μg g^{-1} wet wt.)	References
1974–1989	Baltic	Guillemot (*Uria aalge*)	1974	10	Egg	68[a]	Wideqvist et al. (1993)
			1976	2	Egg	130[a]	
			1978	10	Egg	30[a]	
			1982	10	Egg	25[a]	
			1987	10	Egg	7.7[a]	
			1989	2	Egg	21[a]	
1980s–now	Bear Island, Arctic	Glaucous gull (*Larus hyperboreus*)	1995	3	Liver	<LOD-0.12	Herzke et al. (2003)
			1999	15	Liver	<LOD-0.054	
	Svalbard, Arctic	Glaucous gull (*L. hyperboreus*)	1995	4	Liver	<LOD-0.04	
		Skuas (*Catharacta* sp.)	1993–1994	10	Egg	0.025	Muir et al. (2002) and the references therein
		Penguin (*Pygoscelis* sp.)	1993–1994	10	Egg	0.0011	
	Midway Atoll, Pacific Ocean	Blackfooted albatross (*Diomedea nigripes*)	1994–1995	2	Egg	0.53	Muir et al. (2002)
	Trischen, northern Germany		1981	10	Egg	0.057	Witte et al. (2000)
			1989	10	Egg	0.023	
			1997	10	Egg	0.009	

[a]Concentrations reported on lipid weight basis.

8.1.7. DDT

DDT (1,1,1-trichloro-2,2-bis(*p*-chlorophenyl)ethane) was first synthesized by Othmar Zeidle in Germany in 1874. Its insecticidal properties were not discovered until 1939 by the Swiss chemist Paul Müller. It was widely used in the Second World War to protect the troops against malaria, typhus and other vector borne diseases (Smith, 1991). After the war, DDT was widely used on agricultural crops and in disease vector control (Van Metre et al., 1997).

Technical-grade DDT is a mixture that consists of *p,p′*-DDT (85%) as an active ingredient, *o,p′*-DDT (15%), and *o,o′*-DDT (trace amounts). It also contains DDE (1,1-dichloro-2,2-bis(*p*-chlorophenyl)ethylene)/*p,p′*-DDE and DDD (1,1-dichloro-2,2-bis(*p*-chlorophenyl)ethane)/*p,p′*-DDD as impurities. DDT breaks down into DDE and DDD. The peak production and use of DDT was around 82 million kg in 1962 and at the time DDT was registered for use on 334 agricultural commodities (ATSDR, 2002b). Because of the growing concern about the adverse effects on the environment, especially on wild birds, its production declined abruptly at ~2000 tons in the early 1970s (ATSDR, 2002b). In 1973, all uses of DDT were discontinued in the United States except for emergency public health uses and special cases (ATSDR, 2002b). Before the ban of DDT, more than 80% of DDT was used for agricultural purposes mainly on cotton and followed by peanuts and soybean. *p,p′*-DDD was also used as an insecticide and *o,p′*-DDD is used medically in the treatment of cancer of the adrenal gland, while DDE has no commercial use (ATSDR, 2002b; WHO, 1995). DDT was widely used in China after it was introduced in the 1950s (Li et al., 1999). From 1951 to 1983, the production of DDT was estimated to be 0.27 million tons. Although DDT was banned in 1983, its production for export continues, as does that for dicofol production. DDT is present as an impurity in an acaricide (mitecide) known as Dicofol, which is currently being used widely in China. Dicofol may contain from 1.8% to 2.4% of DDT in the formulation sold in south China (Leung et al., 2005). In China, ~250 metric tons of DDT is currently used annually as an additive in the production of antifouling paint. It is estimated that the accumulative total of DDT used for this purpose since 1950s has reached 10,000 metric tons. This use of DDT is not permitted under the Stockholm Convention, and the Chinese government is taking steps to address this problem.

DDT is not highly toxic to birds when compared with fish. It is best known for the adverse effects on reproduction that is associated with the effects on eggshell thinning, especially DDE. Ratcliffe established the relationship between eggshell thickness and the widespread use of toxic substances (De Luca-Abbott et al., 2001). The decline of the double-crested cormorant population on the Great Lakes of North America in the 1960s and the reduction of the population on Lake Ontario to three pairs were attributed to the breakage of eggs, with 95% breaking before hatching (Weseloh et al., 1983). Cooke et al. (1976) demonstrated a clear linear relationship between the proportion of pair breaking eggs and DDE. However, there is a large discrepancy between the concentrations of DDE that is required to induce critical eggshell thinning, even though in the same species (Fitzner et al., 1988; Henny et al., 1984; Thomas & Anthony, 1999).

In Britain, the levels of *p,p′*-DDE that were found in the eggs of shags between 1963 and 1971, and in those from Ireland in 1964 are shown in Table 8.7. It is considered to be much lower than the levels that were found in the eggs of terrestrial predators during the same period (Walker, 1990). There was no decline in the levels of DDE in shag eggs between 1963 and 1971. The DDE levels of sandwich terns from the Netherlands ($<1\,\mu g\ g^{-1}$ wet wt.) were lower than those that were found in Great Britain in the 1960s. In contrast, very high levels of *p,p*-DDE were found in gannet eggs from Bonaventure Island, Canada between 1968 and 1970. In other studies of the Mediterranean and the Black Sea between 1980 and 1984, high levels of *p,p*-DDE were found in some locations, such as the Danube delta, Majorca and the Po delta (Walker, 1990).

The levels of total DDT in certain bird species were available in Asia between 1993 and 2000 (Table 8.7). The highest levels of total DDT were found in common cormorants in Shinobuza, Japan in 1993 (Guruge et al., 1997) and very high total DDT levels were found in white-tailed sea-eagles in Hokkaido, Japan in 1997 (Kunisue et al., 2003). Little ringed plovers and pond herons from India in 1995, and white-breasted water hen that were collected in Vietnam in 1997 also had high levels of total DDT (Minh et al., 2002; Tanabe et al., 1998). Somewhat lower levels of total DDT were found in little egrets and black-crowned night herons in Hong Kong in 2000 (Connell et al., 2003). In contrast, the egg collected from Lake Tai, China in 2000, of black-crowned night herons, little egrets and thick-billed reed warblers had higher total DDT levels than those of Hong Kong. However, the total DDT levels were lower in Bombay ducks in Jinshan, China between 2000 and 2001 (Nakata et al., 2005). The lowest levels of DDT were found in green back herons and gray herons that were collected in the Philippines in 1994 and in Lake Baikal, in Russia in 1996 and 1997, respectively (Kunisue et al., 2002, 2003).

In Antarctica, low levels of total DDT were found in the eggs of penguins in 1995 and 1996 (Corsolini et al., 2006). In contrast, the levels of total DDT or *p,p′*-DDE that were found in murre eggs in the Arctic region were higher (Braune et al., 2001; Vander Pol et al., 2004).

Table 8.7. DDT concentrations in bird samples from different countries

	Location	Species	Year	*n*	Tissue	Concentration (μg g^{-1} wet wt.)	References
1960s–1980s	Britain	Shag (*Phalacrocorax aristolles*)	1963–1971	n.s[a].	Egg	0.55–3.1	Walker (1990) and the references therein
		Sandwich tern (*S. sandvicensis*)	1963–1965	n.s.	Egg	0.2–1.1	
	Ireland	Shag (*P. aristolles*)	1964	n.s.	Egg	0.3–0.7	
	Netherlands	Sandwich tern (*S. sandvicensis*)	1965–1966	n.s.	Egg	0.61	
	Bonaventure Island, Canada	Gannet (*Sula bassana*)	1968	n.s.	Egg	28 (17–50)	
			1969	n.s.	Egg	31 (19–50)	
			1970	n.s.	Egg	34 (22–57)	
	Mediterranean and the Black Sea						
	Danube delta	Common cormorant (*P. carbo*)	n.s.	n.s.	Egg	19–57	Walker (1990) and the references therein
	Majorca	Corys shearwater (*Calomectris diomedea*)	1984	n.s.	Egg	13–19	
	Po delta	Little tern (*Sterna albifrons*)	1980–1982	n.s.	Egg	5.8–11	
		Gull-billed tern (*S. nilotica*)	1980–1982	n.s.	Egg	4.7–12	

	Japan						
1990–now	Shinobuza	Common cormorant (*P. carbo*)	1993	8	Liver	13	Guruge et al. (1997)
	Hokkaido	White-tailed sea-eagle (*Haliaeetus albicilla*)	1997	2	Liver	1.5	Kunisue et al. (2003)
	India	Little ringed plover (*Chladrius dubius*)	1995	5	Whole body	4.3	Tanabe et al. (1998)
		Pond heron (*Ardeola grayii*)	1995	2	Whole body	3.5	
	Vietnam	White-breasted waterhen (*Amaurornis phoenicurus*)	1997	3	Whole body	3.8	Minh et al. (2002)
	China						
	Hong Kong	Little egret (*E. garztta*)	2000	9	Egg	1.2	Connell et al. (2003)
		Night heron (*Nycticorax nyticorax*)	2000	9	Egg	0.6	
	Lake Tai	Little egret (*E. garztta*)	2000	36	Egg	0.41	Dong et al. (2004)
		Night heron (*N. nyticorax*)	2000	65	Egg	1.1	

Table 8.7. (*Continued*)

Location	Species	Year	*n*	Tissue	Concentration (μg g^{-1} wet wt.)	References
Lake Tai	Thick-billed reed warbler (*Acrocephalus aedon*)	2001	3	Whole body	5.1	Nakata et al. (2005)
Jinshan	Bombay duck	2001	2	Whole body	1.6	
Philippines	Green backed heron (*Butrides striatus*)	1994	3	Whole body	0.058	Kunisue et al. (2003)
Lake Baikal, Russia	Grey heron (*Ardea cinerea*)	1996	2	Breast muscle	0.027	Kunisue et al. (2002)
Alaska	Murre (*Uria aalge*)	1999–2000	67	Egg	0.12	Vander Pol et al. (2004)
Ross sea, Antarctica	Adelie penguin (*Pygoscelis adeliae*)	1995–1996	6	Egg	0.023	Corsolini et al. (2006)
Canadian Arctic	Black-legged kittiwake (*Rissa tridactyla*)	1998	15	Egg	0.06	Braune et al. (2001)
	Thick-billed murre (*U. lomvia*)	1998	15	Egg	0.1	
	Northern fulmur (*Fulmarus glacialis*)	1998	15	Egg	0.21	

1975–1998	Canadian Arctic	Northern fulmur (*F. glacialis*)	1975	15 (pooled samples)	Egg	0.67	Braune et al. (2001)
			1976	12 (pooled samples)	Egg	0.86	
			1977	15 (pooled samples)	Egg	0.43	
			1987	6 (pooled samples)	Egg	0.22	
			1993	15 (pooled samples)	Egg	0.4	
			1998	15 (pooled samples)	Egg	0.21	
1975–1998	Canadian Arctic	Black-legged kittiwake (*Rissa tridactyla*)	1975	12 (pooled samples)	Egg	0.24	Braune et al. (2001)
			1976	6 (pooled samples)	Egg	0.42	
			1987	3 (pooled samples)	Egg	0.11	
			1993	15 (pooled samples)	Egg	0.071	
			1998	15 (pooled samples)	Egg	0.06	
1975–1998	Canadian Arctic	Thick-billed murre (*U. lomvia*)	1975	9 (pooled samples)	Egg	0.20	Braune et al. (2001)
			1976	9 (pooled samples)	Egg	0.23	

Table 8.7. (*Continued*)

Location	Species	Year	*n*	Tissue	Concentration ($\mu g\ g^{-1}$ wet wt.)	References
		1977	9 (pooled samples)	Egg	0.23	
		1987	9 (pooled samples)	Egg	0.16	
		1988	9 (pooled samples)	Egg	0.1	
		1993	15 (pooled samples)	Egg	0.14	
		1998	15 (pooled samples)	Egg	0.1	

[a]n.s.: Not stated.

There were declining trends of the total DDT/*p,p′*-DDE residues in seabird eggs in the Canadian Arctic and the Alaskan Arctic. The levels of total DDT in the eggs of black-legged kittiwakes, northern fulmars and thick-billed murres that were collected in the Canadian Arctic reduced from 1975 to 1998 (Braune et al., 2001).

8.1.8. Polychlorinated biphenyls (PCBs) and Polychlorinated dibenzo-p-dioxins/ dibenzofurans (PCDDs/PCDFs)

8.1.8.1. PCBs

PCBs are mixtures of chlorinated hydrocarbons. Regarding the source of contamination, they can be divided into two main groups: intentional (commercial products) and unintentional (by-products of combustion, such as an incinerator; trace amounts of PCBs have been reported in agricultural chemicals or chemical products as impurities) (UNEP Chemicals, 2002). PCBs were produced commercially in the United States from 1929 to 1977. Due to their chemical inertness, heat resistance, a high dielectric constant and nonflammable properties, they have been used extensively for a variety of industrial purposes, such as dielectrics in transformers and large capacitors as heat exchange fluids and in plastics (WHO, 1993).

They are produced under different trade names, such as Aroclor, Askarel, and Thermino. They are commercially produced as complex mixtures that contain multiple isomers at different degrees of chlorination that are named with a four-digit number. The first two digits refer to the number of carbon atoms in the biphenyl ring (12 for PCBs) and the second two digits indicate the percentage of chlorine by mass in the mixture. The peak production of Aroclor in the United States was in 1970 with a total volume of 85 million pounds (39 million kg) (WHO, 1995). Due to their widespread environmental problems and detrimental health effects, the first set of effluent standards for PCBs was issued by USEPA in 1977 and manufacturing and import limitations of PCBs were issued in 1979. In China, the production of PCBs from 1965 to 1975 was estimated to be 10,000 tons and they were known as PCB3 and PCB5 (Fu et al., 2003). The major uses of PCBs in China were in the electrical industry, such as dielectric fluid in capacitors and transformers, and they were also used in carbon-free copy papers and in paint additives. The uses of PCBs were banned in the 1980s in China.

There are 209 possible PCBs from three monochlorinated isomers to the fully chlorinated decachlorobiphenyl isomer. The toxicology of PCBs depends on the number and position of chlorine atoms. Coplanar PCBs refer to the PCBs without *ortho* substitution, whereas noncoplanar PCBs refer to others (WHO, 1993). Coplanar PCBs have similar effects as dioxins on Ah-receptors in that they bind to the receptor and thus they are usually regarded as dioxin-like compounds (see below). There is much information on the effects of PCB mixtures or congeners on human or laboratory animals and wildlife; however, it is difficult to compare the levels because different studies may report different PCB mixtures or individual congener groups. Similarly, it is not always possible to compare analytical data for PCBs in different studies because different laboratories use different PCB standards or methods of quantification.

Although there are 209 possible PCBs, there are common effects of PCBs on laboratory animals or wildlife. Coplanar PCBs can bind to Ah-receptor that affects defense mechanisms against foreign compounds. PCB residues in eggs were found to be associated with reproductive effects; however, the levels of PCBs that cause this phenomenon varies greatly from species to species. For example, an oral dosage of 1 mg $kg^{-1}day^{-1}$ was linked to lower hatching rates in chickens (*Gallus gallus*), whereas 105 mg $kg^{-1}day^{-1}$were reported to have no effect on the hatching success of mallards (*Anas platrhychos*) (Haseltine & Prouty, 1980). Many studies have concentrated on PCBs and reproductive success, such as hatching success and eggshell thickness. Custer et al. (1999) performed a logistic regression of hatching success with organochlorine levels in double-crested cormorants. They showed that DDE was identified as the major risk factor, not dieldrin and PCBs, and pointed out that many studies focused on PCBs and neglected other more important compounds, such as DDE.

8.1.8.2. Polychlorinated dibenzo-p-dioxins/dibenzofurans (PCDDs/PCDFs)

The PCDDs and PCDFs are two groups of tricyclic planar aromatic compounds. PCDDs and PCDFs are not manufactured commercially except on a small scale for chemical and toxicological research, but they are known to occur naturally, such as in the incomplete combustion of organic material due to forest fires or volcanic activity (ATSDR, 1994b, 1998). PCDDs and PCDFs are produced unintentionally during uncontrolled chemical reactions that involve the use of chlorine, undesired by-products in various chemical formulations, and combustion or incineration processes (ATSDR, 1994b, 1998; WHO 1985). As there can be one to eight chlorine atoms that attach to the benzene rings, they can be categorized into eight families that depend on the degree of chlorination. Hart et al. (1991) investigated the relationship between the concentrations of PCDDs and PCDFs in great blue heron eggs and the effects on chicks. They showed that 10, 135 and 211 ng kg^{-1} wet wt. 2,3,7,8-tetrachlorodibenzo-*p*-

dioxin (or 2,3,7,8-TCDD) in eggs did not cause any effect on the survival of chicks. However, the increased TCDD levels decreased the growth, depressed skeletal growth and increased subcutaneous edema of chicks. Shortened beaks and a scarcity of down follicles in the chicks were also observed from more PCDD- and PCDF-contaminated sites.

To facilitate the evaluation of the toxicity of PCDDs and PCDFs, and also certain PCB congeners, international toxicity equivalency factors (TEFs) are assigned to individual PCDDs or PCDFs and some congeners of PCBs. 2,3,7,8-TCDD is regarded as the most toxic substance among PCDDs, PCDFs and PCBs and it can bind to the Ah-receptor strongly. This receptor is responsible for the control of the mixed function oxidase system, which is an important defense mechanism against foreign compounds. TEFs are assigned to each of the individual dioxins, furans, and coplanar PCBs based on their toxicity relative to 2,3,7,8-TCDD (estimated in terms of their binding affinities to the Ah-receptor). The strength of this binding is used to calculate the TEFs for different dioxins, furans, and PCB congeners with TEF of 2,3,7,8-TCDD set at 1. For each compound, this number is then multiplied by the corresponding concentration to obtain the toxic equivalent (TEQ). This approach can be used in the reverse; the degree of induction of the enzyme is measured and then converted into dioxin equivalents (Safe, 1992).

The TEFs of individual PCDDs, PCDFs and PCB congeners can be obtained from Van den Berg et al. (1998). Usually, the TEQ is calculated from PCDDs, PCDFs and Co-planar PCBs to evaluate whether there is a risk for the tested samples against the value that is obtained from relevant animal tests.

There is a decreasing trend of total PCBs from 1975 to 1998 in Alaska (Table 8.8). Vander Pol et al. (2004) conducted a study on the temporal change of POPs in Alaskan murre (*Uria* spp.) eggs. The total PCBs in thick-billed (*Uria lomvia*) murre eggs that were collected from Prince Leopold Island, in eastern Canada, showed a decreasing trend from 1975 to 1998 (Vander Pol et al., 2004). A decreasing trend of the total PCBs in the livers of adult northern fulmars (*Fulmarus glacialis*) from Prince Leopold Island was observed from 1975 to 1993 (Muir et al., 1999). The decreasing trend of PCBs was also observed in black guillemots in Iceland from 1976 to 1996 (Ólafsdóttir et al., 2005).

The level of total PCBs of common murre eggs in the Arctic regions was higher than those of penguin eggs in Antarctica (Table 8.8) (Corsolini et al., 2006; Vander Pol et al., 2004). Cormorants in Japan had the highest total PCBs and were followed by herons that were collected in Hong Kong (Connell et al., 2003; Guruge et al., 1997; Kunisue et al., 2003). Cormorants that were collected in Romania had lower levels of total

Table 8.8. PCBs concentrations in bird samples from different countries

	Location	Species	Year	*n*	Tissue	PCB method[a]	Concentration (ng g^{-1} wet wt.)	References
1975–1998	Prince Leopold Island, Eastern Canada	Thick-billed murre (*Uria lomvia*)	1975–1977	55	Egg	A	820	Vander Pol et al. (2004) and the references therein
			1987–1988	18	Egg	67	190	Braune et al. (2001)
			1993–1998	30	Egg	67	140	
1975–1993	Prince Leopold Island, Eastern Canada	Northern fulmur (*Fulmarus glacialis*)	1975	7	Liver	A	1100	Muir et al. (1999)
			1976	7	Liver	A	2000	
			1987	8	Liver	A	560	
			1993	10	Liver	A	340	
1990s–now	Canadian Arctic	Common murre (*U.* spp)	1999–2000	67	Egg	46	110	Vander Pol et al. (2004)
	Ross sea, Antarctica	Adelie penguin (*Pygoscelis adeliae*)	1995–1996	6	Egg	46	25	Corsolini et al. (2006)
	Japan							
	Shinobazu pond	Common cormorant (*Phalacrocorax carbo*)	1993	8	Liver	K 300, 400, 500	40,000	Guruge et al. (1997)
	Lake Biwa	Common cormorant (*P. carbo*)	1993	9	Liver	K 300, 400, 500	7900	

Hong Kong	Little egret (*Egretta garztta*)	2000	9	Egg	K 300, 400, 500, 600	960	Connell et al. (2003)
	Night heron (*Nycticorax nyticorax*)	2000	9	Egg	K 300, 400, 500, 600	230	
Romania	Common cormorant (*P. carbo*)	2001	2	Liver	23	110	Covaci et al. (2006)
Philippines	Green backed heron (*Butrides striatus*)	1994	5	Whole body	K 300, 400, 500, 600	85	Kunisue et al. (2003)
Vietnam	Common moorhen (*Gallinula chloropus*)	1997	1	Whole body	K 300, 400, 500, 600	40	Kunisue et al. (2003)
	Cinnamon bittern (*Ixobrychus cinnamomeus*)	1997	1	Whole body	K 300, 400, 500, 600	210	
India	Little egret (*Egretta garzetta*)	1995	1	Whole body	K 300, 400, 500, 600	34	Tanabe et al. (1998)
	Pond heron (*Ardeola grayii*)	1995	2	Whole body	K 300, 400, 500, 600	42	
Russia	Grey heron (*Ardea cinerea*)	1996–1997	2	Breast muscle	K 300, 400, 500, 600	7.7	Kunisue et al. (2002)

[a]Methods used to analyse PCBs were based on Aroclor 1254 standard (A), total number of PCB congeners used and Kanechlor preparations.

PCBs than those from Hong Kong, but were higher than those from the Philippines and Vietnam (Covaci et al., 2006). Green backed herons in the Philippines and common moorhen and cinnamon bitterns in Vietnam had similar levels of total PCBs but the herons in India had lower PCBs levels (Kunisue et al., 2003; Minh et al., 2002; Tanabe et al., 1998). The gray herons in Russia had the lowest level of total PCBs (Kunisue et al., 2002).

The levels of PCDDs and PCDFs in waterbirds that were collected at different periods from different countries were compared (Table 8.9) (Choi et al., 2001; Harris et al., 2003; Kannan et al., 2003; Kubota et al., 2004; Senthilkumar et al., 2002; Wan et al., 2006). Bald eagle muscles that were collected from the Upper Peninsula of Michigan, in 2000, had relatively high levels of PCDDs and PCDFs, whereas the livers of white tailed sea eagles from eastern Germany from 1990 to 1998, the eggs of double-crested cormorants that were collected from Canada between 1990 and 1998, and the livers of various species of birds that were collected in Japan from 1997 to 2001 had relatively low levels of PCDDs and PCDFs. The livers of herring gulls that were collected from China in 2002, the subcutaneous fat of black-tailed gulls that was collected from Korea between 1992 and 1994, and the livers of white-tailed sea eagles from west Poland showed lower levels of PCDDs and PCDFs.

Total PCDDs and PCDFs were usually used for comparison, however, it should be noted that different studies used different standards to calculate total PCDDs/PCDFs. The eggs of double-crested cormorants that were collected from Mandarte Island, and Crofton and Mitlenatch Islands, in the Strait of Georgia, in British Columbia, Canada, between 1973 and 1998 have been analyzed for PCDDs and PCDFs (Harris et al., 2003). The study showed that the levels of total PCDDs and PCDFs in 1998 were lower than those from 1973 to 1989 (Table 8.9). In another study, the PCDDs/PCDFs levels in white-tailed sea eagle livers were analyzed (Kannan et al., 2003). It showed that no significant difference in the total PCDDs/PCDFs in adult females that were collected from 1985 to 1995 was observed. However, this observation was confounded by small sample sizes and associated inherent biological variations.

8.2. Biological effects

Organochlorine pesticides, PCBs and PCDDs/PCDFs caused the decline of several avian populations in the late 1960s, such as sparrow hawks and kestrels in Europe and North America (Newton, 1992). The effects of these compounds on birds were linked with direct mortality due to pollutants and reproductive performance, such as eggshell thinning and

Table 8.9. Polychlorinated dibenzo-*p*-dioxins/ dibenzo-*p*-furans (PCDDs/PCDFs) concentrations in bird samples from different countries

	Location	Species	Year	*n*	Tissue	TEQ (pg of TEQ g^{-1}, lipid wt.)	Concentration (pg g^{-1}, lipid wt. corrected)	References
1990s–now	Upper Peninsula of Michigan	Bald eagle	2000	6	Muscle	21,000	34,000	Senthilkumar et al. (2002)
	Eastern Germany	White-tailed sea-eagle (*Haliaeetus albicilla*)	1990–1998	19	Liver	–	2500	Kannan et al. (2003)
	Strait of Georgia, BC, Canada	Double-crested cormorant (*Phalacrocorax auritus*)	1990–1998	70	Egg	42	1200	Harris et al. (2003)
	Lake Biwa, Japan	Common cormorant (*P. carbo*)	2001	26	Liver	1200	2800	
	Chiba, Tokyo, Japan	Gray heron (*Ardea cinerea*)	1997–1998	2	Liver	1800	11,000	Senthilkumar et al. (2002)
		Spot-billed duck (*Anas poecilorhyncha*)	1997–1998	2	Liver	1500	1500	
		Whimbrel (*Numenius phaeopus*)	1998	1	Liver	400	2200	
		Short-tailed shearwater (*Puffinus tenuirostris*)	1998	1	Liver	18	33	
		Cattle egret (*Bubulcus ibis*)	1999	1	Liver	1800	2700	
		Great egret (*Ardea alba*)	1999	1	Liver	3500	4900	
	Tianjin, China	Herring gull (*Larus argentatus*)	2002	20	Liver	–	860	Wan et al. (2006)

Table 8.9. (*Continued*)

	Location	Species	Year	*n*	Tissue	TEQ (pg of TEQ g^{-1}, lipid wt.)	Concentration (pg g^{-1}, lipid wt. corrected)	References
	Nakdong River estuary, Korea	Black-tailed gull (*L. crassirostris*)	1992–1994	10	Fat	230	390	Choi et al. (2001)
	Western Poland	White-tailed sea-eagle (*Haliaeetus albicilla*)	1996–1998	4	Liver	–	440	
1973–1998	Mandarte Island, Canada	Double-crested cormorant (*Phalacrocorax auritus*)	1973	14 (pooled samples)	Egg	1500	2000	Harris et al. (2003)
			1979	5 (pooled samples)	Egg	1800	3500	
			1985	5 (pooled samples)	Egg	2600	7100	
			1989	20 (pooled samples)	Egg	1800	2800	
			1990	11 (pooled samples)	Egg	1300	2000	
			1991	10 (pooled samples)	Egg	690	880	
			1992	10 (pooled samples)	Egg	1200	1700	
			1994	15 (pooled samples)	Egg	660	870	
			1995	10 (pooled samples)	Egg	1100	1200	
			1998	14 (pooled samples)	Egg	500	730	

decreased reproduction success (Table 8.10, see also De Luca-abbott et al., 2001). In addition, body condition, such as body mass controlled for body size, including head and bill, showed a significant negative relationship with these contaminants, and all organochlorines (e.g. HCB, DDE, PCBs), except oxychlordane in females but not in the males of great black-backed gulls (Helberg et al., 2005) may also affect immune status and function (Bustnes et al., 2004). Significant or near significant positive relationships ($0.1 > p > 0.001$) were found between most persistent organochlorines and the levels of heterophils in the blood for both sexes and the lymphocytes of male gulls (Bustnes et al., 2004). There is evidence that organochlorines affect immune systems, which may decrease their efficiency and make birds more susceptible to parasites and diseases.

Some studies have suggested there is interaction among organochlorine pesticides, PCBs and PCDDs/PCDFs with other compounds (Blus et al., 1983). For example, a toxic interaction between dieldrin and chlordane has been suggested that caused mortality in wild birds (Blus et al., 1983; Okoniewski & Novesky, 1993). Different cyclodienes may have bound to a common target site—a gamma-aminobutyric acid-regulated chloride ion channel in a nerve membrane—that resulted in convulsions (Cole & Casida, 1986). The additive effects of chlordane and dieldrin have been demonstrated in rats (Keplinger & Deichmann, 1967). Other interactive effects among organochlorine pesticides and PCBs have been discussed elsewhere (Stansley & Roscoe, 1999; Walker, 1990).

8.3. Limitations

There are important limitations in understanding the spatial and temporal trend of the pollutant levels of waterbirds, which is particularly true in China. Many studies that have been conducted in China have focused on water and sediment. There is limited information of pollutant levels in waterbirds available in China. In addition, a number of studies have investigated target pollutants with different tissue samples of different species. It has been determined that different tissue samples of species of birds can accumulate varying degrees of pollutants. It would be more instructive to compare tissue samples within species. Moreover, it is difficult to adequately compare current results with previous ones (i.e. 1940s–1970s) because of the advanced capability and improved sensitivity of instruments, and the application of different extraction methods and standards for quantification.

Standard protocol that uses the same tissue of birds is useful to evaluate the global level of contaminants, spatially and temporally. The egg is

Table 8.10. Adverse effects caused by organochlorine pesticides, PCBs and TCDD

Compounds	Species	Means of dosage/tissue measured	Mean concentration in tissue measured/dosage	Adverse effect	References	Remarks
Heptachlor epoxide	American Kestrel (*Falco sparverius*)	Dietary fed	$\geqslant 1.5\,\mu g\ g^{-1}\ day^{-1}$	Reproductive impairment	Henny et al. (1983)	
	Canada geese (*Branta canadensis*)	Dietary fed	$\geqslant 10\,\mu g\ g^{-1}\ day^{-1}$	Reproductive impairment	Blus et al. (1984)	
	Canada geese (*B. canadensis*)	–	$\geqslant 10\,\mu g\ g^{-1}\ day^{-1}$	Nest success lower		
Dieldrin	Japanese quail (*Coturnix coturnix japonica*)	Dietary fed	$20\,mg\ kg^{-1} day^{-1}$	Reduced survival of parent, reduced egg/hen	WHO (1989) and the references therein	9 weeks
		Dietary fed	$30\,mg\ kg^{-1} day^{-1}$	Reduced survival of parent, reduced egg/hen, reduced hatchability, reduced survival of chick		7 weeks
		Dietary fed	$40\,mg\ kg^{-1} day^{-1}$	Reduced survival of parent, reduced egg/hen, reduced survival of chick		6 weeks
	Bobwhite quail (*Colinus virginianus*)	Dietary fed	$10\,mg\ kg^{-1} day^{-1}$	Reduced survival of parent		34 weeks
		Dietary fed	$20\,mg\ kg^{-1} day^{-1}$	Reduced survival of parent, reduced egg/hen		34 weeks

		Dietary fed	40 mg $kg^{-1}day^{-1}$	Reduced survival of parent, reduced egg/hen		34 weeks
HCB	Japanese quail (*C. coturnix japonica*)	Dietary fed	100 µg g^{-1} day^{-1}	Increased mortality	WHO (1995) and the references therein	90 days
			20 µg g^{-1} day^{-1}	Reduced hatchability		
TCDD	Three-day-old chickens	Dietary fed	1 or 10 µg g^{-1} day^{-1}	Suffered from subcutaneous and pulmonary edema, distended abdomens and increased peritoneal and pericardial fluid	Hart et al. (1991) and the references therein	3 weeks
Mirex	Chicken	Dietary fed	300 mg $kg^{-1}day^{-1}$	Reduced survival of hatchlings	Davison et al. (1975) and the references therein	12 weeks
		Dietary fed	600 mg kg^{-1}	Reduced survival of hatchlings and hatchling survival		6 weeks
	Mallard (*Anas platyrhynchos*)	Dietary fed	100 mg kg^{-1}	Egg shell thinning and reduced duckling survival	Waters et al. (1977)	
	American Kestrel (*F. sparverius*)	Dietary fed	8 mg kg^{-1}	Reduced sperm concentration and increased semen volume	Bird et al. (1983)	69 days
Toxaphene	Duckling	Dietary fed	10 or 50 mg $kg^{-1}day^{-1}$	Reduced growth and backbone impairment	Mehrle et al. (1979)	90 days

Table 8.10. (*Continued*)

Compounds	Species	Means of dosage/tissue measured	Mean concentration in tissue measured/ dosage	Adverse effect	References	Remarks
p,p′-DDE	Black-crowned night heron (*Nycticorax nycticorax*)	Eggs	8200*	Decreased fledging success	Henny et al. (1984)	
p,p′-DDE	Common cormorant (*Phalacrocorax carbo*)	Eggs	4000	Eggshell thinning	Dirksen et al. (1995)	
p,p′-DDE	Little egret (*Egretta garzetta*)	Eggs	1000	Young survival	Connell et al. (2003)	
p,p′-DDE	Double-crested cormorant (*Phalacrocorax auritus*)	Eggs	960–11,000	Decreased reproduction	Custer et al. (1999)	
p,p′-DDE	Dalmatian Pelican (*Pelecanus crispus*)	Eggs	14,000–22,000	Eggshell thinning	Crivelli et al. (1989)	
			Mean concentration of dosage/in tissue measured			
p,p′-DDE	Great Egret (*Ardea alba*)	Liver	124,300	Shell breakage	Pratt (1972)	
p,p′-DDE	Great Blue Heron (*A. herodias*)	Eggs	> 10,000	Reproductive impairment	Custer et al. (1998)	
p,p′-DDE	Great Blue Heron (*A. herodias*)	Eggs	3000	Reduced hatching	Blus (1996)	

p,p′-DDE	Great Blue Heron (*A. herodias*)	Liver	569,740	Lethal effects	Call et al. (1976)
$\sum$PCBs	Common cormorant (*P. carbo*)	Liver	40,000	Sublethal effects	Guruge et al. (2000)
$\sum$PCBs	Common cormorant (*P. carbo*)	Liver	319,000	Adult mortality	Koeman et al. (1973)
$\sum$PCBs	Common cormorant (*P. carbo*)	Eggs	7300–8200	Unhatched eggs, Deformed bill	Larson et al. (1996)
$\sum$PCBs	Double-crested cormorant (*P. auritus*)	Eggs	3500	Reproductive success	Tillitt et al. (1992)
$\sum$PCBs	Double-crested cormorant (*P. auritus*)	Eggs	6600–7300	Chick deformities	Yamashita et al. (1993)
$\sum$PCBs	Double-crested cormorant (*P. auritus*)	Eggs	30,000	Embryo mortality	Barron et al. (1995)
$\sum$PCBs	Black-crowned night heron (*N. nycticorax*)	Eggs	10,000–63,000	Decreased hatching and fledging success	Sakellarides et al. (2006) and the references therein

a better option for the biomonitoring of pollutants, and its advantages and limitations in biomonitoring have been discussed (De Luca-abbott et al., 2001; Peakall, 1994). Basically, pollutants can be transferred from a female to her eggs, hence, the measurement of pollutant levels in eggs can provide an idea of the level of pollutants in the environment that are bioavailable to birds. It can also reduce confounding factors from birds, such as sex, age and feeding habits. However, the availability and accessibility of the eggs of some birds make it difficult for routine use in biomonitoring under certain circumstances.

8.4. Study on persistent organic pollutants in local waterbirds in Hong Kong

Among the wildlife in Hong Kong, waterbirds are one of the conspicuous groups of animals in our coastal environment, and they have high conservation value in the Hong Kong marine ecosystem. In Hong Kong, waterbirds are mainly found in two estuaries, namely Deep Bay and Starling Inlet (Carey, 1998; Young & Cha, 1995). In recent years, there is increasing evidence that the western waters are under threat from a wide range of environmental contaminants as compared to the eastern part of Hong Kong. A complete evaluation of the environmental impacts of these pollutants to the Hong Kong coastal environment would be instructive to provide a scientific basis for risk characterization and management.

Waterbirds are valuable/useful for environmental monitoring. Waterbirds are particularly susceptible to environmental contaminants because they are long-lived and are top trophic-level animals in the food web. Consequently, they are able to integrate pollutant levels over a broad area by bioaccumulation (Furness, 1993). Contaminant levels in waterbird eggs, therefore, provide important and useful information for monitoring changes in the environmental quality of the Hong Kong coastal areas. This is particularly relevant in the Northwestern part of Hong Kong, which is heavily influenced by increased activities arising from the rapid industrialization of the PRD region. In Hong Kong, Ardeids (Herons and Egrets) represent the dominant and the most conspicuous groups of resident waterbirds. Local Ardeid species include Chinese Pond Heron (*Ardeola bacchus*), Cattle Egret (*Bubulcus ibis*), Little Egret (*Egretta garzetta*), Great Egret (*Egretta alba*) and Black-crowned Night Heron (*Nycticorax nycticorax*).

A study commissioned by the Agriculture, Fisheries and Conservation Department (AFCD) of Hong Kong, "Study on the effect of water pollution on the breeding success of Ardeids" completed in 2001, detected organochlorines in the eggs of Little Egrets (*Egretta garzetta*) and Black-crowned

Night Herons (*Nycticorax nycticorax*) collected from Mai Po and A Chau, respectively (Lam et al., 2001). The results indicated that Little Egret and Black-crowned Night Heron eggs had concentrations of DDE (a metabolite of DDT) sufficient to initiate adverse effects on the breeding success of these species (Connell et al., 2003). In addition, total PCBs present in eggs were at threshold levels where adverse effects could be initiated with the Little Egret, but not Black-crowned Night Herons.

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