Chapter 10
Organochlorine Pesticide Residues in Foodstuffs, Fish, Wildlife, and Human Tissues from India: Historical Trend and Contamination Status

V. Dhananjayan and B. Ravichandran

Abstract Ever since people started utilizing natural resources, environmental quality started to deteriorate. Deterioration of the quality of these resources affects human health and well-being and therefore becomes a threat to human security. Organochlorine pesticides (OCPs) are ubiquitous environmental contaminants relevant due to their high toxicity and potential carcinogenicity. These contaminants are considered to be hazardous to aquatic organisms, fish, birds, and humans. Varying levels of these pesticides have been reported in different segments of the ecosystem including humans. Health damage to fish and wildlife has prompted concern about the health effects of these contaminants on humans. It has been found that a greater amount of total intake of these contaminants in human beings is through consumption of contaminated food. A number of abnormalities seen in the reproductive system of various wildlife species can be correlated with similar abnormalities on the rise in the human population. Exposure to these pollutants also suppresses the immune system, thereby increasing the risk of acquiring several diseases. Temporal trends examined by comparing the results of previous studies on OCP levels in the Indian environment revealed a decline in the trend of dichlorodiphenyltrichloroethanes (DDTs) and hexachlorocyclohexanes (HCHs) in some parts of the natural environment. In contrast, very high concentrations were detected in biotic samples. Continuous monitoring and epidemiological studies of OCP levels in humans are warranted. In this chapter, we outline the environmental and human health problems associated with pesticide contamination. To our knowledge, this is the first report to present the residue levels of persistent OCPs in fish, wildlife, and human tissues from India.

Keywords Organochlorine pesticides · Exposure · Effects · Fish · Wildlife · Human

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10.1 Introduction

The application of pesticides to agriculture has greatly improved the food production worldwide. India is the second largest producer of vegetables after China and accounts for 13.4 % of world production. Surveys carried out by institutions spread throughout the country indicate that 50–70 % of vegetables are contaminated with insecticide residues. India has a wide variety of climate and soils on which a range of vegetable crops can be grown (Karanth 1982). During the last three decades, considerable emphasis has been laid on production of crops in India and vegetable exports have been stepped up. However, increased use of chemical pesticides has resulted in contamination of the environment and also caused many associated long-term effects on human health (Bhanti and Taneja 2007). The presence of pesticide residues in food commodities has always been a matter of serious concern. The problem is especially serious when these commodities are consumed (Solecki et al. 2005). Pesticides have been associated with a wide spectrum of human health hazards, ranging from short-term impacts such as headaches and nausea to chronic impacts like cancer, reproductive harm, and endocrine disruption (Berrada et al. 2010). The heavy use of pesticides may result in environmental problems like disturbance of natural balance, widespread pest resistance and environmental pollution, hazards to non-target organisms, wildlife, and humans.

India is among the largest agricultural societies in the world as the agricultural sector provides livelihood to the majority of its 1 billion people. Modern agriculture uses inputs such as chemical fertilizers, pesticides, seeds of high-yielding varieties, and mechanization that resulted in increased yields ushering an era of green revolution in the country. Synthetic pesticides are one of the major agro inputs that significantly contributed to the agricultural production in the country. Pesticides may have helped in enhancing agricultural production, but at the same time these chemicals have caused adverse effects (Shetty and Sabitha 2009).

Organochlorine pesticides (OCPs) are an important potential component of chemical pollutants used extensively for agriculture and public purposes in India as these are comparatively cheap and effective. These persistent organic compounds such as hexachlorocyclohexanes (HCHs) and dichlorodiphenyltrichloroethanes (DDTs) are the predominant chemical contaminants found in various environmental matrices in India. Our biggest concern is that these molecules are stable in the environment. It is suspected that most of water bodies and soils are contaminated with these chemicals or with their degradation products (Krisahnamurthy 1984). HCH, aldrin, dieldrin, and heptachlor are banned in India. However, gamma-HCH (lindane) and DDT have restricted use and are allowed for termite control and public health purposes, respectively. Greater concentrations of organochlorine (OC) residues in human breast milk and adipose tissues (Kutz et al. 1991) collected from various developing countries indicated increased exposure of humans in these regions to OC insecticides. In more developed nations, foodstuffs are monitored through periodical surveys to assess human exposure and to maintain public health standards (Hotchkiss 1992; Krieger et al. 1992; Yess et al. 1993). However, comprehensive nationwide residue monitoring studies are prohibitively expensive and are yet to be implemented in many developing nations. Despite the continuing use of OCs, little information is available on their levels in foodstuffs in India.

An international convention aiming to restrict persistent organic pollutants (POPs) was formally adopted in May 2001, and global actions to reduce and eliminate release of the pollutants have been recommended. The OCPs form part of the "dirty dozen" and have already had a strong impact on wildlife and human beings (Choi et al. 2001). Chlorinated pesticides have shown to give rise to estrogen-dependent reproductive effects in several avian species (Fry 1995); further, DDT and its metabolites have also been shown to cause obstruction of Ca^{2+} metabolism in birds (Lundholm 1994). However, for the clarification of the toxic effects of OCs, basic information such as the distribution of OCs among organ (or tissue) is essential.

There are 234 pesticides registered in India. Out of these, 4 are WHO Class Ia pesticides, 15 are WHO Class Ib pesticides, and 76 are WHO Class II pesticides, together constituting 40 % of the registered pesticides in India. In terms of consumption, too, the greatest volumes consumed are of these poisons. India is the fourth largest pesticide producer in the world after the USA, Japan, and China. During 2003–2004, the domestic production of pesticides was approximately 85,000 t, and about 60,000 t was used annually (Anonymous 2005) against 182.5 million ha of land where 70 % accounts for DDTs, HCHs, and organophosphate pesticides (Bhattacharyya et al. 2009; Nirula and Upadhyay 2010). The domestic consumption of pesticides in agriculture is comparatively low (0.5 kg/ha; only 3.75 % of global consumption) against 12.0, 7.0, 6.6, and 3.0 kg/ha in Japan, USA, Korea, and Germany, respectively (Chauhan and Singhal 2006). Our objective in this chapter is to provide a comprehensive account of the distribution of OCPs in foodstuffs, fish, wildlife, and human tissues from India: historical trend and contamination status and their environmental sources, their movement through the food chain, and possible ecotoxicological risk of health in biota including humans.

10.2 Historical Trend and Contamination Status

The birth of modern pesticide era was hailed as a major breakthrough for mankind. The persistent pesticides belonging to the group of OC compounds were potent weapons against vectors of diseases, pests of crops, forests, and rangeland. Originally, certain pesticides like DDT and HCH were imported in the formulated form for mosquito control; however, slowly India started using these pesticides for agricultural purposes as well. The large-scale manufacture and distribution of OCPs did not take place until after the accidental discovery of DDT as an insecticide by Muller in 1946. Later, while several countries banned DDT, India stopped its use in agriculture in 1989. However, DDT is still being manufactured and used for malaria control. Pesticide demand is close to 90,000 t per annum. Insecticides

(73 %) dominate the market, followed by herbicides (14 %) and fungicides (11 %). Cotton, rice, and wheat growers account for almost 70 % of pesticide consumption, and the states consuming more are Andhra Pradesh, Punjab, Karnataka, and Gujarat. Presently, 44 types of pesticides are manufactured in India. DDT, benzene hexachloride (BHC), malathion, and carbofuran are commonly used. About 83,000 t of pesticides are used in the agricultural sector annually (Menon 2003). About 144 pesticide molecules are registered in India and 65 technical-grade pesticides are manufactured indigenously (The Pesticides Scenario 2000).

In India, in 1960, about 600 t of DDT was used in agriculture and 21,000 t for public health. As against this for public health in 1980, DDT used was 15,500 t and 9,100 t of DDT was used for 1983–1984. With increased use of the chemicals world over, negative environmental effects began to be noticed. These included toxicity in non-target organisms, bioaccumulation, long-term environmental persistence, and persistence in target pests. Since its introduction in Indian agriculture, about 575,000 t of BHC have been used—500,000 t in agriculture and 75,000 t in the public health sector to date. The current annual consumption of HCH is nearly 30,000 t in agriculture and 6,000 t in public health. It forms 60 % of the total insecticides consumed in the agriculture sector (Gupta 1986). Effects of pesticide use on ecosystems in India have scarcely been investigated.

Though complete ban on DDTs was imposed in many developed nations, in India it was banned only for agricultural use, but it is still used for malaria control, and HCH also is not completely banned (still used for agriculture). About 31 banned or restricted pesticides in other countries are still in use in India and about 350,000 t since 1985 and 7,000 t in 2001–2002 of DDT were used (http://www.cseindia.org/2002). Earlier studies in Indian marine environment showed that polychlorinated biphenyls (PCBs) and OCP residues are comparable with those found in other developing countries, but suggested an increase in the future due to the continuing usage of OCPs in agriculture and vector control measures. HCH and DDT were introduced in 1948 and 1949, respectively, for agricultural and public health purposes. Their cumulative consumption until 1995 was estimated to be 500,000 t for DDT and 1 million t for HCH. The values suggest that India has been the major consumer of HCH in the world (Kannan et al. 1995). Until 1992, these two insecticides accounted for two-thirds of the total consumption of pesticides in the country (Kannan et al. 1992a).

In global comparison, HCH levels in India are 1–2 orders of magnitude higher than those in countries like Cambodia (Kunisue et al. 2004b), Philippines (Kunisue et al. 2002), Vietnam (Minh et al. 2004), and Indonesia (Sudaryanto et al. 2006), but relatively lower than those in China (Kunisue et al., 2004a) and Hong Kong (Wong et al. 2002). The levels reported in Chennai by Subramanian et al. (2007) were among the highest found so far. It is also worthy to note that high levels of HCHs were also found in various environmental and biotic samples from India (Senthilkumar et al. 2001; Kunisue et al. 2003; Ramu et al. 2007) suggesting that India acts as a major source for HCHs which can also contribute to pollution by these compounds (Table 10.1).

Table 10.1 Top ten countries with highest technical HCH use (Li et al. 1999)

Number	Country	Total usage (kt)
1	China	4,464
2	India	1,057
3	Soviet Union (Erstwhile)	693
4	France	520
5	Egypt	479
6	Japan	400
7	USA	343
8	Germany East	142
9	Spain	133
10	Mexico	132

10.3 OCPs in Foodstuffs

Many surveys invariably indicated the predominance of DDT and HCH in Indian foodstuffs. Residues of chlorinated pesticide (OCP) in food have given rise to major concerns. This has reflected in the large number of reports in the literature on this subject. Moreover, the chronic effects of such exposure levels from food intake are mostly unknown but there is growing evidence of carcinogenicity and genotoxicity as well as endocrine disruption capacity (Miller and Sharpe 1998) being attributed to the ingestion of or exposure to pesticides. Despite the fact that the use of certain OCPs in agriculture is prohibited in many countries, these compounds have been detected in the environment worldwide due to their persistent nature (Rejendran and Subramanian 1999).

The long persistence of some agrochemicals in the environment sets in a series of undesirable effects through contamination of food and feed. Food contamination surveys have been conducted in this country since the late 1960s, when the Insecticide Act was enacted in 1968. Nevertheless, most of these studies have been conducted by certain institutions with a limited number of market samples. Report on comprehensive nationwide monitoring has not as yet been performed. The results of some of the earlier surveys have been reviewed by Kalra and Chawla (1981). Bioaccumulation of pesticides and biomagnification processes have become the weak links in the food chain. Among the pesticides that have acquired notoriety, DDT and HCH are particularly important. Although now partially banned, they are still very much in use because of their wide spectrum of activity and ready availability at low cost (Krisahnamurthy 1984; Aslam et al., 2013). During the last few decades, widespread contamination and toxic effects of organic chemicals have become a serious environmental problem. These chemicals enter the soil by direct treatment or being washed off from the plant surface during rainfall. Their physicochemical characteristics, which include hydrophobicity and resistance to degradation, cause these chemicals to accumulate in soils and sediments (Hong et al. 2008; Hu et al. 2010). Soil and sediments can act as contributors of organic pollutants to the atmosphere, especially of semi-volatile compound in warm climates. The fate of pesticides in soils with different cropping land use has been extensively studied worldwide including India (Viet et al. 2000; Oldal et al. 2006; Senthil Kumar et al. 2009).

10.3.1 Vegetables

Pesticides are widely used to ensure high crop yields. Indian diet contains vegetables as important component in food, because majority of Indians are vegetarian and per capita consumption of vegetables is approximately 135 g/day. Therefore, information on pesticide residues in vegetables is very important for human health. Bankar et al. (2012) investigated pesticide residues in market foods in Uttar Pradesh, India. About 58.33 % of the samples were free from residues, 28.33 % of samples contained pesticide residues at or below maximum residue limit (MRL), and 13.33 % of samples contained pesticide residues above MRL. Brinjal was the most positive followed by cabbage, tomato, and lady's finger. A study conducted by Mukherjee et al. (2011) in West Bengal, India, to access the level of OCPs residues in vegetables revealed that the concentration of total OCPs (ΣOCPs) ranged between < 0.01 and 65.07 μg/kg with an average of 9.67 ± 2.34 μg/kg (wet wt.). The concentration of total DDTs (ΣDDTs), total HCHs (ΣHCHs), aldrin, dieldrin, and heptachlor was 3.49 ± 0.93 μg/kg, 2.07 ± 0.53 μg/kg, 1.32 ± 0.65 μg/kg, 1.36 ± 1.18 μg/kg, and 1.80 ± 0.4 μg/kg (wet wt.), respectively.

Bakore et al. (2002) conducted a study during 1993–1996 to investigate the magnitude of contamination of OC insecticides in vegetables which were brought for sale to the consumers in the local markets of Jaipur City, Rajasthan, India. Samples of vegetables (potato, tomato, cabbage, cauliflower, spinach, and okra) collected at the beginning, middle, and end of the seasons with respect to different vegetables and OC levels were assessed. Most of the collected samples were found to be contaminated with residues of DDT and its metabolites (dichlorodiphenyldichloroethane (DDD) and dichlorodiphenyldichloroethylene (DDE) isomers of HCH (alpha, beta, and gamma-HCH), heptachlor, heptachlor epoxide, and aldrin. Some of the detected insecticides exceeded the limit of tolerance prescribed by World Health Organization (WHO)/Food and Agriculture Organization (FAO). Kumari et al. (2002) monitored pesticide contamination among market samples (60) of six seasonal vegetables during 1996–1997. The estimation of insecticide residues representing four major chemical groups, i.e., OC, organophosphorous, synthetic pyrethroid (SP), and carbamate, was done. The tested samples showed 100 % contamination with low but measurable amounts of residues. Among the four chemical groups, the organophosphates were dominant followed by OCs, SPs, and carbamates. About 23 % of the samples showed contamination with organophosphorous compounds above their respective MRL values. More extensive studies covering different regions of Haryana state are suggested to get a clear idea of the magnitude of vegetable contamination with pesticide residues.

Kumari et al. (2004) analyzed 84 farm gate samples of seasonal vegetables for pesticide residues. About 26 % samples contained residues above MRL values. The contamination was mainly with organophosphates followed by SPs and OCs. The residues of HCH, DDT, and endosulfan were found in all the samples but did not exceed the tolerance limit. Bhanti and Taneja (2005) analyzed the summer and winter vegetable samples during 2002–2003 for pesticide residue estimation. The contamination levels of winter vegetables (average concentration of 4.57, 6.80, and

5.47 ppb, respectively, for lindane, endosulfan, and DDT) were found to be slightly higher than those of the summer vegetables (average concentration of 4.47, 3.14, and 2.82 ppb, respectively, for lindane, endosulfan, and DDT). The concentrations of these OCPs in summer and winter vegetables were well below the established tolerances but continuous consumption of such vegetables even with moderate contamination level can accumulate in the receptor's body and may lead to chronic effects that could be fatal.

Bhanti and Taneja (2005) assessed the pesticide contamination in vegetables grown in different seasons (summer, rainy, and winter). Data obtained were then used for estimating the potential health risk associated with the exposure to these pesticides. The pesticide residue detected in vegetables of different seasons shows that the winter vegetables are the most contaminated followed by summer and rainy vegetables. The concentrations of the various pesticides were well below the established tolerances but continuous consumption of such vegetables even with moderate contamination level can accumulate in the receptor's body and may prove fatal for human population in the long term. The analysis of health risk estimates indicated that chlorpyrifos and malathion did not pose a direct hazard; however, exposure to methyl parathion has been found to pose some risk to human health.

A recent study by Kumar et al. (2010) investigated the OCP residues in five varieties of vegetable samples collected from different markets in Uttar Pradesh. Vegetables tested in this study do not contain any quantities of pesticide residues hazardous to humans. Kumar et al. (2012) analyzed pesticides, namely, aldrin, dieldrin, heptachlor, and lindane in selected root and leaf vegetables collected from Kolkata. The concentration of total OCPs ranged between <0.01 and 6.00 ng/g, with an average of 2.16 ± 0.21 ng/g (wet wt.). The concentration of individual aldrin, dieldrin, heptachlor, and lindane was 0.48 ± 0.06 ng/g, 0.13 ± 0.02 ng/g, 1.03 ± 0.11 ng/g, and 0.52 ± 0.06 ng/g (wet wt.), respectively. The selected vegetables had residue levels much below the recommended MRLs set by the European Commission and the Indian Government (Tables 10.2 and 10.3).

Kumar et al. (2011) have conducted a study to assess the levels of OCPs (DDTs, HCHs, and endosulfan) in vegetables from Kolkata, India. The total concentration of OCPs ranged between 0.29 and 106.65 $\mu g\ kg^{-1}$ (wet wt.). The results indicated that all the vegetable samples had some levels of one or more OCPs. The mean concentration of DDT, HCH, and endosulfan was 6.63 ± 1.17 $\mu g\ kg^{-1}$ (wet wt.), 4.29 ± 1.47 $\mu g\ kg^{-1}$ (wet wt.), and 1.34 ± 0.99 $\mu g\ kg^{-1}$ (wet wt.), respectively. The ratio of α-HCH to γ-HCH isomers (α/γ-HCH ratio) ranged from 0.03 to 5.69, which reflects the use of lindane as well as technical formulation of the HCH. The ratios of (DDE + DDD)/ΣDDT and DDT/DDE were 0.52 and 1.21, respectively, which indicate that these vegetables were contaminated with fresh input as well as biotransformation of DDTs. Residue levels of HCH, DDT, and endosulfan in vegetables from Kolkata market were below the MRLs set by the European Commission and the Indian Government indicating minimal risk to the consumers.

Srivastava et al. (2011) conducted a study on 20 vegetables to analyze 48 pesticides including 13 OCs, 17 organophosphates (OPs), 10 SPs, and 8 herbicides (H). A total number of 60 samples, each in triplicates, were analyzed using a quick, easy,

Table 10.2 MRLs (European Commission 2006) for OCPs in the selected fruits and vegetables

Commodity	MRLs (mg/kg)						
Fruits	Gamma-HCH	Methoxychlor	Aldrin	Dieldrin	Endrin	*p, p'*-DDE	DDT
Papaya	0.01	0.01	0.01	0.01	0.01	0.05	0.05
Water melon	0.01	0.01	0.03	0.03	0.01	0.05	0.05
Banana	0.01	0.01	0.01	0.01	0.01	0.05	0.05
Mango	0.01	0.01	0.01	0.01	0.01	0.05	0.05
Pear	0.01	0.01	0.01	0.01	0.01	0.05	0.05
Pineapple	0.01	0.01	0.01	0.01	0.01	0.05	0.05
Vegetables							
Tomato	0.01	0.01	0.01	0.01	0.01	0.05	0.05
Lettuce	0.01	0.01	0.01	0.01	0.01	0.05	0.05
Cabbage	0.01	0.01	0.01	0.01	0.01	0.05	0.05
Carrot	0.01	0.01	0.01	0.01	0.01	0.05	0.05
Onion	0.01	0.01	0.01	0.01	0.01	0.05	0.05
Cucumber	0.01	0.01	0.02	0.02	0.01	0.05	0.05

Table 10.3 MRLs for pesticides in vegetables

OCP compounds (ng/g)					
MRLs	Aldrin	Dieldrin	Heptachlor	Lindane	References
Europe	10	10	10	50	European Commission 2008
India	100	100	50	1,000	FSSAI 2011

FSSAI Food Safety Standards Authority of India

cheap, effective, rugged, and safe method. About 23 pesticides were detected from total 48 analyzed pesticides in the samples with the range of 0.005–12.35 mg/kg. The detected pesticides were above MRL (PFA 1954). However, in other vegetables the level of pesticide residues was either below detection limit (BDL) or MRL.

Kumari et al. (2012) monitored 80 winter vegetable samples during 1997–1998 for pesticidal contamination. All the tested samples were contaminated with pesticide residues with measurable amounts. Among the four major chemical groups, residue levels of organophosphorous insecticides were the highest followed by bicarbamates, SPs, and OCs. About 32% of the samples showed contamination with organophosphorous and carbamate insecticides above their respective MRL values. These studies suggested that more extensive monitoring studies covering all vegetable crops from different agro-climatic regions of the state be carried out to know the exact level of pesticidal contamination, which may serve as a basis for future policy on chemical use.

10.3.2 Tea

Tea is a perennial plantation crop grown under monoculture providing favorable conditions for a variety of pests. The concept of pest control has undergone a considerable change over the past few decades. In recent years, there has been a greater dependence on the use of pesticides (7.35–16.75 kg/ha) with little importance laid on other safe control methods for the management of tea pests. Due to this practice, the tea pests showed a higher tolerance/resistance status due to formation of greater amount of esterases, glutathione S-transferase, and acetylcholinesterase. Thus, over-reliance on pesticides ends up with pesticide residue in made tea (DDT—10.4–47.1 %; endosulfan—41.1–98.0 %; dicofol—0.0–82.4 %; ethion—0.0–36.2 %; cypermethrin—6.0–45.1 %). The growing concern about the pesticide residue in made tea, its toxicity hazards to consumers, the spiraling cost of pesticides, and their application have necessitated a suitable planning which will ensure a safe, economic, as well as effective pest management in tea. At present, it is a global concern to minimize chemical residue in tea and the European Union and German law imposed stringent measures for the application of chemicals in tea and fixed MRL values at ≤ 0.1 mg/kg for the most commonly used pesticides, which is not met in the real practice and has been a major constraint to tea-exporting countries like India. In order to regulate the situation of the Indian market at a global level, central insecticide board and prevention of food adulteration regulation committee have reviewed the MRL position for tea and has recommended 10 insecticides, 5 acaricides, 9 herbicides, and 5 fungicides for use in tea and issued the tea distribution and export control order in 2005 which will help the country to limit the presence of undesirable substances in tea (Gurusubramanian et al. 2008).

Bishnu et al. (2009) quantified the residues of organophosphorus (e.g., ethion and chlorpyrifos), OC (e.g., heptachlor, dicofol, endosulfan), and SP (e.g., cypermethrin and deltamethrin) pesticides in made tea, fresh tea leaves, soils, and water bodies from selected tea gardens in the Dooars and hill regions of West Bengal, India, during April and November, 2006. The organophosphorus (OP) pesticide residues were detected in 100 % substrate samples of made tea, fresh tea leaves, and soil in the Dooars region. In the hill region, 20–40 % of the substrate samples contained residues of organophosphorus (OP) pesticides. The OC pesticide residues were detected in 33–100 % of the substrate samples, excluding the water bodies in the Dooars region and 0–40 % in the hill region. The estimated mean totals of studied pesticides were higher in fresh tea leaves than in made tea and soils. The SP pesticide residues could not be detected in the soils of both the regions and in the water bodies of the Dooars. Sixteen percent and 20 % of the made tea samples exceeded the MRL level of chlorpyrifos in Dooars and hill regions, respectively. Based on the study, it was revealed that the residues of banned items like heptachlor and chlorpyrifos in made tea may pose health hazards to the consumers.

10.3.3 Dry Fruits

The use of pesticides on cash crops and exportable food commodities had always been a serious concern. Fruits form one of the important constituents of human diet, in that they give one-third of the requirement of calories, vitamins, and minerals. Kumari et al. (2005) have reported the pesticide residues in butter (45) and ghee (55) samples collected from rural and urban areas of cotton growing belt of

Haryana. Butter samples were comparatively more contaminated (97 %) than ghee (94 %), showing more contamination with OC insecticides from urban samples. About 11 % samples of butter showed endosulfan residues above MRL value and 2 % samples had residues of SPs and organophosphates each above their respective MRL values. In ghee, residues of HCH and DDT both and of endosulfan exceeded the MRL values in 5 and 20 % samples, respectively. Among organophosphates, only chlorpyriphos was detected with 9 % samples showing its residue above MRL value. Irrespective of contamination levels, residues above the MRL values were more in ghee. More extensive study covering other agricultural regions/zones has been suggested to know the overall scenario of contamination of milk products. Pandey et al. (2010) have carried out a study to determine the OCP residues in commonly used dry fruits like cashew nut, walnut, coconut, chilgoza, *chironji, makhana,* resins, apricot, almonds, date palm, and pistachio nut collected from local markets of Lucknow, India. The results indicate the presence of very low levels of HCH (0.007–1.328 mg kg^{-1}), DDT (BDL–0.140 mg kg^{-1}), and endosulfan (BDL–0.091 mg kg^{-1}). There are no MRL values established for nuts in the country.

10.3.4 Diet

Battu et al. (2005) analyzed 46 samples each of vegetarian and nonvegetarian total diet consumed from March 1999 to December 2002 by male subjects in the age group of 19–24 years to assess their risk through dietary intake with respect to pesticide residues. The results revealed low dietary intake of levels of DDT which were almost comparable to levels reported in developed countries. The results are indicative of contamination of total diet with pesticide residues despite a ban on the use of DDT and restricted use of lindane in agriculture only. Predominance of lindane residues indicates that liquid milk was a main contributory source as it constitutes almost 21 % of the total diet consumed per day. Concerted efforts by regulatory authorities and emphasis on judicious use of agrochemicals in pest control are required to decrease the burden of these chemicals in food stuffs to levels safe for dietary intake.

10.3.5 Bovine Milk and Dairy Products

In a multicentric study that assessed the pesticide residues in selected food commodities collected from different states of our country, *p, p'*-DDE was found in 82 % of the study samples of bovine milk collected from 12 states (Toteja 1993). Pandit et al. (2002) monitored the milk and dairy product samples of various brands from different cities in Maharashtra, India, to determine the presence of OCP residues contamination. Trace levels of DDT and HCH were detected in the samples. Total HCH levels in milk and milk product samples were lower than total DDT levels, which could be attributed to earlier extensive antimalaria sanitary activities.

Butter had higher levels of DDT than cheese and milk powder. All levels of OCP residues in milk and milk products were well below the maximum permissible limits given by the FAO/WHO.

The risk posed by the presence of OCPs in milk and milk products was estimated for the population. Pandit and Sahu (2002) determined the levels of OCPs in milk and milk products in and around Mumbai City. A total of 520 samples were used to determine the mean daily consumption of milk and milk products by different age groups and these data were used to evaluate the daily exposure to the public. Non-cancer effects were evaluated by comparing the predicted exposure distributions to the published guidance values. For chemicals identified as potential human carcinogens, cancer risk was evaluated using standard methodology. The majority of the chlorinated pesticides identified in the milk and milk product samples studied were found to be at levels which do not pose unacceptable risks to the public, with the exception of HCH. The cancer risk estimated for this chemical slightly exceeds the US Environmental Protection Agency (EPA) guidance value.

Indian Council of Medical Research (2001) has reported DDT residues in 82 % of the 2,205 samples of bovine milk collected from 12 states. About 37 % of the samples contained DDT residues above the tolerance limit of 0.05 mg/kg (whole milk basis). The highest level of DDT residues found was 2.2 mg/kg. The proportion of the samples with residues above the tolerance limit was maximum in Maharashtra (74 %) followed by Gujarat (70 %), Andhra Pradesh (57 %), Himachal Pradesh (56 %), and Punjab (51 %). In the remaining states, this proportion was less than 10 %. Data on 186 samples of 20 commercial brands of infants' formulae showed the presence of residues of DDT and HCH isomers in about 70 % and 94 % of the samples with their maximum level of 4.3 and 5.7 mg/kg (fat basis), respectively.

John et al. (2001) conducted a survey during 1993–1996 to investigate the magnitude of contamination of bovine milk with OCP residues from Jaipur City, Rajasthan, India. Milk samples, i.e., dairy (toned and whole) and buffalo milk, were collected seasonally, and pesticide residues were assessed. The results indicate that all the milk samples were contaminated with DDT and its metabolites, and isomers of HCH, heptachlor, and its epoxide, and aldrin. Seasonal variations of these pesticide residue levels were also observed in all the milk samples. Samples collected during winter season were found to contain higher residue levels as compared to other seasons.

Monitoring of bovine milk of different places in Bundelkhand region of India was carried out by Nag and Raikwar (2008) to evaluate the status of OCP residues. Out of a total of 325 samples, 206 (63.38 %) were contaminated with residues of different OCPs. The average concentration of total HCH was 0.162 mg/kg. Among the different HCH isomers, the frequency of occurrence of α-isomer was the highest followed by δ, γ, and β. Endosulfan (α, β, and sulfate) was detected in 89 samples with a mean concentration of 0.0492 mg/kg, while total DDT comprising DDT, DDE, and DDD was present in 114 samples having a mean concentration of 0.1724 mg/kg.

One hundred forty-seven samples of bovine milk were collected from 14 districts of Haryana, India, during December 1998 to February 1999 and analyzed for the presence of OCP residues. ΣHCH, ΣDDT, Σendosulfan, and aldrin were detected in 100, 97, 43, and 12 % samples and with mean values of 0.0292, 0.0367, 0.0022, and 0.0036 μg/ml, respectively. Eight percent samples exceeded the MRL of 0.10 mg/kg as recommended by WHO for ΣHCH, 4 % samples of 0.05 mg/kg for α-HCH, 5 % samples of 0.01 mg/kg for γ-HCH, 26 % samples of 0.02 mg/kg for β-HCH as recommended by PFAA, and 24 % samples of 0.05 mg/kg as recommended by FAO for ΣDDT. Concentrations of β-HCH and *p, p′*-DDE were more as compared to other isomers and metabolites of HCH and DDT (Sharma et al. 1999).

10.3.6 Fish

Singh and Singh (2008) investigated the ΣHCH and ΣDDT, aldrin, endosulfan, and chlorpyrifos in liver, brain, and ovary, gonadosomatic index (GSI), and plasma levels of testosterone (T) and estradiol-17β (E2) during breeding season of captured catfishes and carps from the unpolluted ponds of Gujartal, Jaunpur (reference site) and polluted rivers Gomti, Jaunpur and Ganga, Varanasi. Results have indicated that catfishes have higher bioaccumulation of pesticides than the carps: it was beyond the permissible limits for ΣHCH, whereas for ΣDDT only in catfishes of polluted rivers. The GSI and plasma levels of T and E2 were lowered in the fishes captured from the polluted rivers. It was concluded that the fishes of Gomti and Ganga reflect the degree of pesticide pollution present in those water bodies.

Muralidharan et al. (2009) determined the OCP residues in ten species of fishes caught at Cochin and Rameshwaram coast and sold in Coimbatore, Tamil Nadu, India. Species were selected on the basis of their regular availability throughout the year and commercial value. A total of 389 fishes were analyzed for OC residues and their suitability for human consumption was evaluated. Results show varying levels of residues of HCH, DDT, heptachlor epoxide, endosulfan, and dieldrin. About 22 % of the fishes exceeded the MRLs of total HCH prescribed by FAO/WHO for fish products. The calculated dietary intake of total HCH through consumption of *Carangoides malabaricus, Chlorophthalmus agassizi,* and *Sardinella longiceps* exceeded the maximum acceptable daily intake (ADI) limits prescribed for human consumption. The present study recommends continuous monitoring of environmental contaminants in marine fishes to assess the possible impact on human health.

Dhananjayan and Muralidharan (2010b) assessed the contamination status of inland wetlands of India, through evaluating the OCP residues in fishes collected from different inland wetlands in Karnataka, India and their suitability for human consumption. Among the OCPs tested, isomers of HCH were the most frequently detected with β- and γ-HCH as the main pollutants. Average concentration of ΣHCH and ΣDDT ranged from 2.1 to 51.7 lg/kg and BDL to 12.3 lg/kg, respectively. Other OCPs such as heptachlor epoxide, dieldrin, and endosulfan were found at lower levels. OCPs detected in the present study were well below the tolerance limits recommended for fishes. The calculated daily dietary intake of OCPs in all the species examined was lower than the maximum ADI limits prescribed for human consumption (Table 10.4).

Table 10.4 Comparison of calculated dietary intake concentration of OCPs with the ADI stipulated by various statutory agencies

OCPs	The average calculated dietary intake concentration through consumption of fishes (µg/person/day consumption; Dhananjayan and Muralidharan 2010b)	Allowable limits by various statutory agencies (µg/person/day)
ΣHCH	0.61	18[a]
HE	0.21	–
ΣEndosulfan	0.15	450[b]
Dieldrin	0.13	6[a]
ΣDDT	0.27	300[b]

[a] Health Canada (1996)
[b] IARC (International Agency for Research on Cancer 1989)

Dhananjayan et al. (2012a) evaluated the OCP residues in the inland wetland fishes of Gujarat, India and their suitability for human consumption. Among the various OCPs analyzed, γ-HCH and β-HCH were detected in 70–80 % of samples. *p, p′*-DDE, the metabolite of DDT, was detected in higher load. DDT and HCH detected in the present study were well below the tolerance limits recommended for fishes. The calculated daily dietary intake of OCPs in all the species examined was lower than the maximum ADI limits prescribed for human consumption. Further studies on continuous monitoring of OCPs and dietary intake are warranted to characterize the residue accumulation and to facilitate the early identification of risks due to fish consumption.

10.3.7 Birds and Wildlife

The OCs due to their persistent nature remain stored in the body fat of birds and largely disturb calcium metabolism and thereby induce eggshell thinning over a period of time eventually leading to population decline (Tanabe et al. 1998). The effects of pesticide on wildlife, especially raptors, waterfowl, and fish-eating birds have been extensively studied around the world (Stickel et al. 1969; Fleming et al. 1984). Although food abundance and quality greatly impact reproductive effort and success in many predatory birds (Korpinaki and Norrdahl 1991; Rohner 1996), there has been increasing concern about the potential deleterious effects of OC and other pesticides on the population of many species of birds (Gard et al. 1995; Block et al. 1995). Although reduction of the food supply, disruption of delicate predator–prey relationship, alteration of habitat, and a variety of similar changes could all affect the distribution and abundance of wildlife, the role of pesticide cannot be ruled out.

The pesticide exposure pattern of birds in India presents a varied picture compared to their counterparts in Europe and America. Due to the human and environmental risks associated with the use of such pesticides, they have been banned in several countries but are still used in India and their presence was reported in various studies (Mathew 1993; Saiyed et al. 1999; Kannan et al. 1997). A large number of studies have focused on the accumulation of pesticides in various species of flora and fauna around the world (Minh et al. 2002). There is little information available on the presence of few OCP residues in the tissues of a few species of birds and the higher accumulation of OC in resident and migratory birds from South India (Tanabe et al. 1998). Reports also show that the decline in the population of many species of birds was mainly due to the application of aldrin and monocrotophos in agricultural fields. Declining population of birds breeding at regular sites and infrequent sightings or total absence of insectivorous birds, such as drongos and bee-eaters in the agricultural fields in various parts of India are being reported.

Declining breeding population of Sarus Crane (*Grus antigone*) (Vijayan 1991; Muralidharan 1993; Pain et al. 2004) in Keoladeo National Park, Bharatpur, the reports on the unsuccessful breeding of Himalayan Grey-headed Fishing Eagle, *Ichthyophaga nana,* at Corbett National Park in Uttarakhand (Naoroji 1997), the higher accumulation of OC in resident and migratory birds from South India (Tanabe et al. 1998), fast-disappearing common bird "House Sparrow" in India (Vijayan 2003), and accumulation of various concentration of OCPs in tissues of vultures (Muralidharan et al. 2008) give credence to the concern of ornithologists. In the case of Sarus Crane in Bharatpur, it was inferred that the decline in population was mainly due to application of aldrin in the agricultural fields around the park; 18 Sarus Cranes were found dead inside the park within a span of 3 years due to aldrin (Muralidharan 1993). Even after the ban on aldrin, death of 15 Sarus Cranes due to monocrotophos poisoning was reported (Pain et al. 2004). Unfortunately, mortality of Sarus Crane has become rather frequent. Although such definite information is not available for many species of birds in India, study conducted elsewhere gives definite proof for the deleterious effects of pesticides on avifauna and the ecosystem.

In India, although we keep guessing that pesticide could be the reason for the decreasing population trend in many insectivorous and piscivorous birds, nothing has been proved till date (Muralidharan 2002). Except a very few studies on the problems of pesticide contamination in Indian wildlife and birds, especially resident birds (Muralidharan 1993; Tanabe et al. 1998; Senthil Kumar et al. 1999; Muralidharan et al. 2008), not much of research has been carried out to bring out the exact picture of pesticide contamination in a specific region

10.3.8 Impact of Pesticides on Birds in India

In India, unfortunately systematic monitoring has never been done to evaluate the level of residues and their biological effects. Limited surveys were carried out by various authors that have been mainly confined to very small number of individuals to a particular place and period. The results of scattered studies in the Indian environment are reviewed and discussed as follows: HCH and DDT residues of the internal body organs, depot fat, and blood plasma of a few species of Indian wild birds from Lucknow were estimated by Kaphalia et al. (1981). High levels of DDT were detected in depot fat of crow, kite, and vulture (50.8, 67.0, and 95.3 ppm, respectively) Total HCH detected in depot fat of crow was 29.7 ppm; lesser amounts were found in vulture, kite, and cattle egret, respectively. A single specimen of pigeon ovary examined contained 1.21 ppm total HCH and 0.31 lindane. Kaphalia et al. (1981) reported that within the same food habit group, smaller individuals are likely to ingest larger amounts of pesticide residues. Studies also clearly indicate that, HCH levels in liver, lung, and kidney were generally high in pigeon and crows and in the breast muscles and spleen of vulture. More lindane and total HCH was found in tissues of vulture compared with other species. Avian species, thus, reflect biological magnification of HCH and DDT residues, presumably due to their food habits.

A study by Misra (1989) in monitoring and surveillance of pesticide pollution of Mahala water reservoir with reference to its avifauna reported relatively high levels of total OCP residues in Flamingo and Red-wattled Lapwing brain, as compared with other tissues. Flamingo contained 7.39 µg/g beta HCH and 7.46 µg/g *p, p′*-DDE in brain while Red-wattled Lapwing had 3.45 µg/g alfa HCH, 5.82 µg/g gamma-HCH, and 3.66 µg/g aldrin in brain. Large Pied Wagtail had 5.42 µg/g *p, p′*-DDE in the brains which is much higher than the concentrations present in other species of birds studied. Total DDT residue concentrations were 2.0, 5.42, and 6.34 µg/g in brain of Black-winged Stilt, Large Pied Wagtail, and Flamingo, respectively. In other tissues, total residues of DDT generally occurred in the following order: Large Pied Wagtail > Flamingo > Black-winged Stilt > Indian Sandgrouse > Red-wattled Lapwing. Snipe had no detectable total DDT residues. Total DDT detected was 3.73, 10.53, 0.56, 0.15, and 5.05 µg/g in liver of Black-winged Stilt, Large Pied Wagtail, Indian Sandgrouse, Red-wattled Lapwing, and Flamingo, respectively (Misra 1989).

Misra (1989) found high correlations of OCPs between breast muscle and body tissue when the data from all the individual birds were combined and subjected to a correlation test. The total OC residue levels in brain, alimentary canal, kidney, liver, and ovary correlate at the 0.01 significant levels. Brain, alimentary canal, and kidney rank 1, of which alimentary canal and kidney have the highest correlation (Misra 1989). Muralidharan et al. (1992) conducted a study on OC residues in the eggs of selected colonial water birds breeding at Keoladeo National Park, Bharatpur, India. The study recorded higher concentration of dieldrin in eggs of Large Cormorant (1.54 ppm), Indian Shag (2.94 ppm), Darter (1.52 ppm), Grey Heron (5.95 ppm), Cattle Egret (2.52 ppm), Painted Stork (5.78 ppm), and Spoon Bill (1.3 ppm). As the concentration in all the eggs was higher than 1 ppm, it was suggested that the concentration would have been associated with reproductive impairment (Stickel 1973).

The breeding population of Sarus Crane in Keoladeo National Park in Rajasthan has come down to six pairs as against 27 pairs in 1973 (Walkinshaw 1973). As diagnostic and circumstantial evidence prove the cause of death of 18 Sarus Crane between 1987–1988 and 1989–1990 to be aldrin poisoning (Muralidharan 1993), the reason for the decline in the breeding population becomes obvious. Brain tissue of Sarus Cranes, Collared Doves, and Blue Rock Pigeons collected from Keoladeo National Park, Bharatpur, showed an average of 19.33, 15.19, and 20.42 ppm of dieldrin, respectively. Dieldrin in other tissues ranged from 0.78 to 92.26 ppm in Sarus Cranes, 3.44 to 66.17 ppm in Collared Doves, and 16.92 to 20.99 ppm in Blue Rock Pigeons. Very high residues of aldrin in the gastrointestinal tract (89.75 ppm) and dieldrin at much higher quantities in the brain than the lethal level (4–5 ppm) clearly indicate that dieldrin after being metabolized from aldrin was responsible for the death of many Sarus Cranes in Bharatpur (Muralidharan 1993). Himalayan Grey-headed Fishing Eagle *I. nana* an endangered species has been breeding unsuccessfully for the last 5 years in Corbett National Park in Uttarakhand. Detection of high levels of DDT, HCH, dieldrin, endosulfan, and heptachlor in the unhatched egg (Rishad 1995) makes the reason all the more strong. Unsuccessful breeding of Himalayan Grey-headed Fishing Eagle at Corbett National Park in Uttarakhand was also reported by Naoroji (1997).

Higher accumulation of OC in resident and migratory birds from South India was documented by Tanabe et al. (1998), and Senthilkumar et al. (1999, 2001) measured levels of pesticides, dioxins, and PCBs in a few species of birds. Tanabe et al. (1998) suggest that generally the OC contamination levels in individual species of birds may vary with feeding habits and the residue pattern of OC in most species of resident birds analyzed followed the order of HCHs > DDTs > PCBs. The concentrations of HCHs in resident birds of South India were in the range of 14–8,800 ng/g followed by DDTs ranging in concentrations from 0.3 to 3,600 ng/g. Global comparison of OC concentrations indicated that resident birds in India had the highest residues of HCHs and moderate to high residues of DDTs.

Among the various OC residues analyzed in the tissue of six species of birds collected from Nilgiri District, Tamil Nadu, high levels of endosulfan residues were detected in the tissues of Large Cormorant with values of 10.9, 126.8, 112.88, 217.2, and 233 ppm in brain liver, kidney, muscle, and food content, respectively (Muralidharan and Murugavel 2001). While Muralidharan (1993) attributed the mortality of 18 Sarus Cranes and a few granivorous birds in Keoladeo National Park, Bharatpur to aldrin, Muralidharan et al. (2008) reported the levels of persistent OCP residues in tissues of White-backed Vulture from different locations in India. The levels of DDT, HCH, dieldrin, and endosulfan residues among other OCs have been detected in the tissues and eggs; none of them was indicative of either food chain buildup or poisoning leading to population decline.

Muralidharan et al. (2008) determined OCP residues in tissues of five Indian White-backed Vultures and two of their eggs collected from different locations in India. All the samples had varying levels of residues. *p, p′*-DDE ranged between 0.002 µg/g in muscle of vulture from Mudumali and 7.30 µg/g in liver of vulture from Delhi. Relatively higher levels of *p, p′*-DDT and its metabolites were documented in the bird from Delhi than other places. Dieldrin was 0.003 and 0.015 µg/g, while *p, p′*-DDE was 2.46 and 3.26 µg/g in eggs 1 and 2, respectively. Dieldrin appeared to be lower than the threshold level of 0.5 µg/g. *p, p′*-DDE exceeded the levels reported to have created toxic effects in eggs of other wild birds. Although varying levels of DDT, HCH, dieldrin, heptachlor epoxide, and endosulfan residues were detected in the vulture tissues, they do not appear to be responsible for the present status of population in India.

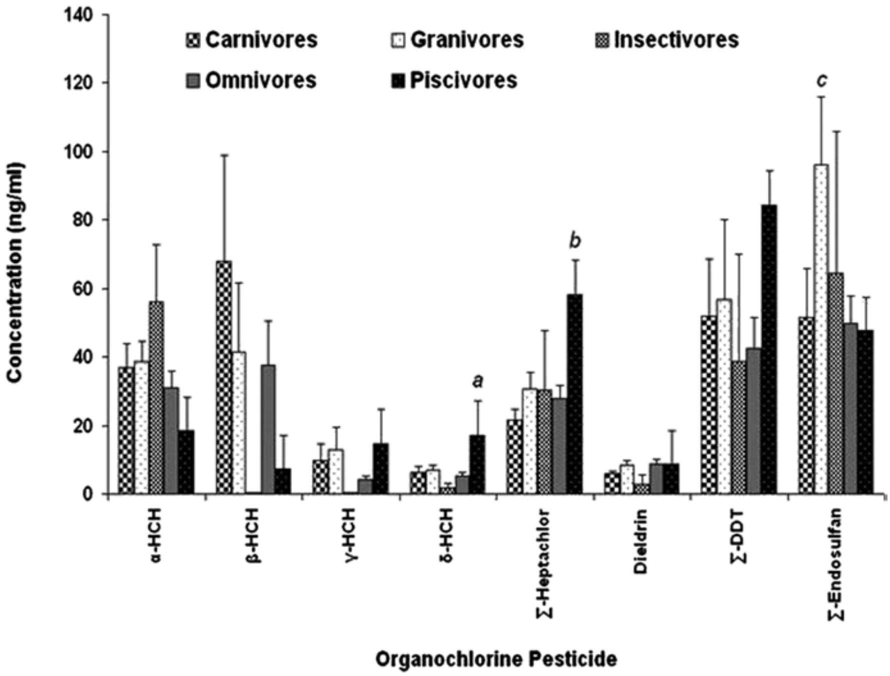

Fig. 10.1 Distribution pattern of OCP residues (mean ± standard error, ng/ml) in plasma of birds according to their feeding habits. $a = P < 0.05$ vs carnivores, granivores, omnivores, and insectivores; $b = P < 0.05$ vs carnivores, granivores, omnivores, and insectivores; $c = P < 0.05$ vs omnivores. (Source: Dhananjayan and Muralidharan et al. 2010a)

Dhananjayan and Muralidharan (2010a) reported the concentrations of OCPs in blood plasma of 13 species of birds collected from Ahmedabad, India. Among the various OCPs determined, HCHs and their isomers had higher contribution to the total OCPs. Concentration of HCHs varied from 11.4 ng/mL in White Ibis (*Threskiornis melanocephalus*) to 286 ng/mL in Sarus Crane (*Grus antigone*), while DDT ranged between 19 ng/mL in Black Ibis (*Pseudibis papillosa*) and 147 ng/mL in Painted Stork (*Mycteria leucocephala*). *p, p′*-DDE was accounted for more than 50 % of total DDT in many of the samples analyzed. However, a *p, p′*-DDT to *p, p′*-DDE ratio higher than 1 obtained for many species of birds indicates the recent use of DDT in this study region. The concentrations of cyclodiene insecticides, heptachlor epoxide, dieldrin, and total endosulfan ranged from 15.8 to 296.2 ng/mL, BDL to 15, and 41.1–153.2 ng/mL, respectively. The pattern of total OCP load generally occurred in the following order: granivores\insectivores\omnivores\ piscivores\carnivores. Although the OC residues detected in blood plasma of birds are not indicative of toxicity, the presence of residues in birds over the years 2005–2007 indicates continued exposure to OC compounds (Fig. 10.1; Table 10.5).

Dhananjayan et al. (2011b) determined the presence of persistent OCPs and PCBs in blood plasma of White-backed Vulture (*Gyps bengalensis*), Egyptian

Table 10.5 OCP residues (ng/ml) in plasma of birds in India

Region	Species	Year	Tissue	*N*	*p, p'*-DDE	dieldrin	Source
India	House Crow	1980	Plasma	3	35 (20–40)	–	Kaphalia et al. 1981
India	Pariah Kite	1980	Plasma	3	100 (43–190)	–	Kaphalia et al. 1981
India	White-backed Vulture	1980	Plasma	3	183 (106–250)	–	Kaphalia et al. 1981
India	Cattle Egret	1980	Plasma	3	29 (26–30)	–	Kaphalia et al. 1981
India	Blue Rock Pigeon	1980	Plasma	3	4 (3–10)	–	Kaphalia et al. 1981
India	House Crow	2005–2007	Plasma	3	34.2 (4.6–51)	4.1 (2.9–6.3)	Dhananjayan and Muralidharan 2010
India	Pariah Kite	2005–2007	Plasma	51	15.3 (<1–137)	9.1 (<1–68.5)	Dhananjayan and Muralidharan 2010
India	Cattle Egret	2005–2007	Plasma	3	38.6 (6.5–101)	3 (<1–7.9)	Dhananjayan and Muralidharan 2010
India	Blue Rock Pigeon	2005–2007	Plasma	34	13 (<1–101)	7.8 (<1–37.7)	Dhananjayan and Muralidharan 2010

Vulture (*Neophron percnopterus*), and Griffon Vulture (*Gyps fulvus*) collected from Ahmedabad, India. All the samples had varying levels of OCPs and PCBs. The mean concentration of HCHs, DDTs, and PCBs among plasma ranged from 43.7 to 136, 8.8 to 64.8, and 226 to 585 ng/ml, respectively. Among the various OCPs analyzed, *p, p'*-DDE was detected most frequently. The concentrations of cyclodiene insecticides detected were lower than the other OC residues. The levels of pesticides measured in plasma samples of three species of vulture were comparable to the results documented for a number of avian species and were lower than those reported to have deleterious effects on survival or reproduction of birds. Although no threat is posed by any of the OCPs detected, continuous monitoring of breeding colonies is recommended. This study is also the first account of a comprehensive analysis of toxicants present in blood plasma of vulture species in India. The values reported in this study can serve as guidelines for future research in general as well as control values during the analysis of samples obtained from birds in the event of suspected OC poisoning (Table 10.6).

Dhananjayan et al. (2011c) reported the information on the current status of contamination by OCPs in eggs and tissues of House Sparrow, *Passer domesticus*, in Tamil Nadu, India. The mean concentration of ΣHCH and ΣDDT in eggs ranged from 0.01 to 1.81 µg/g and 0.02 to 1.29 µg/g, respectively. Concentration of

Table 10.6 Organochlorine chemical residues (ng/ml) in blood plasma of three species of vultures from India. (Source: Dhananjayan et al. 2011b)

	Egyptian vulture (n=6)				Griffon vulture (n=8)				White-backed vulture (n=17)			
	Mean	Min	Max	%	Mean	Min	Max	%	Mean	Min	Max	%
α-HCH	3.67	BDL	9.07	50	38.1	BDL	75.6	38	41.7	BDL	115	56
β-HCH	41.4	BDL	79.1	67	5.9	BDL	11.3	63	83.0[a]	BDL	661	69
γ-HCH	32.1	BDL	127	33	BDL	BDL	BDL	0	6.51	BDL	60.6	50
δ-HCH	BDL	BDL	BDL	0	BDL	BDL	BDL	0	5.62	BDL	16.7	44
ΣHCH	76.56	BDL	215	83	43.7	BDL	86.9	75	136[a]	BDL	770	94
HE	14.4	BDL	17. 9	50	12.9	BDL	14.4	50	23.87	BDL	73.4	75
Dieldrin	4.8	BDL	7.26	33	5.65	BDL	6.34	25	6.51	BDL	15.7	50
p, p′-DDE	8.75	BDL	25.6	83	16.2	BDL	26.5	75	16.28	BDL	39.5	94
p, p′-DDD	6.78	BDL	9.37	33	19.9	BDL	39.3	63	17.58	BDL	163	44
p, p′-DDT	5.42	BDL	8.24	67	3.45	BDL	8.62	38	31.56[a]	BDL	261	63
ΣDDT	8.75	BDL	25.6	83	35.9	26.5	45.2	100	64.83	BDL	455	88
α-Endosulfan	21.21	BDL	25.8	33	BDL	BDL	BDL	0	BDL	BDL	BDL	0
β-Endosulfan	BDL	BDL	BDL	0	27.3	BDL	54	25	8.54	BDL	28	56
Endosulfan sulfate	5.16	BDL	10.1	50	25.8	BDL	39	63	23.31	BDL	76.1	63
ΣEndosulfan	26.24	BDL	33.1	67	52.8	BDL	93	75	31.69	BDL	104	69
ΣPCBs	225.5	186	275	100	253	164.5	342	100	585	109	3,776	100

p, p′-DDE ranged from BDL to 0.64 µg/g, representing more than 60 % of the PDDTs. About 28 % of samples had *p, p′*-DDE levels above the critical concentration associated with reproductive impairment. However, the mean concentrations of cyclodiene insecticides were less than 0.5 µg/g.

A review by Dhananjayan et al. (2011a) explained the exposure and effects of orgnochlorine pesticides in birds in India. Although we have information on the levels of persistent environmental contaminants in many species of birds in India, due to lack of adequate data, we are unable to make any direct correlation with the reported population decline. Of all the birds, levels of DDT, HCH, and dieldrin recorded in some of the species were indicative of poisoning. Although varying levels of many persistent pesticides have been documented, the levels were not indicative of either food chain buildup or poisoning. However, the residue levels recorded were higher than the concentration reported in some other parts of the world. Further, even though very high levels of lead, indicative of poisoning, was observed, it cannot account for the massive reduction that the species has witnessed. However, such levels have to be viewed with concern.

Dhananjayan (2012a) reported that the OCPs and PCBs are responsible for the mortality of waterbirds in Nalaban bird sanctuary in Chilika Lake. One or more residues were detected in all the tissues of birds analyzed. Concentrations of HCHs, DDTs, and PCBs ranged from BDL to 811 ng/g, BDL to 1,987 ng/g, and BDL to 1,027 ng/g, respectively. PCBs levels were less than the food and drug administration's (FDA's) action limits. However, the need for additional research is heightened when considering that some of the birds are classified as a globally protected species by the international bodies.

Dhananjayan (2012b) assessed the persistent OCPs in various tissues of House Sparrow, *Passer domesticus,* from Tamil Nadu, India, between 2001 and 2006. The major compounds quantified in eggs, liver, brain, and muscle tissues are HCH, *p, p′*-DDE, dieldrin, and heptachlor epoxide. The mean concentrations of total polychlorinated biphenyls (ΣPCBs), ΣHCH, and ΣDDT in eggs are 0.94 ± 0.66 μg/g and 0.35 ± 0.26 μg/g on wet wt. basis. Concentrations of *p, p′*-DDE, a metabolite of *p, p′*-DDT, contributed 77 % to the total DDT. Twenty-eight percent of egg samples exceeded the *p, p′*-DDE concentration which was proposed as critical levels for birds.

Dhananjayan (2013) assessed the presence of OCPs in liver tissues of 16 species of birds collected from Ahmedabad, India, during 2005–2007. The higher concentrations of total OCPs were detected in livers of Shikra (*Accipiter badius;* 3.43 ± 0.99 μg/g wet wt.) and the lower levels in White Ibis (*Pseudibis papillosa;* 0.02 ± 0.01 μg/g wet wt.). Concentrations of DDT and its metabolites, HCH and isomers, dieldrin, and heptachlor epoxide were lower than the concentrations reported for various species of birds in India. Accumulation pattern of OCPs in birds was, in general, in the order HCH > DDT > heptachlor epoxide > dieldrin.

10.3.9 Human Blood

OC insecticide residues, especially DDT and HCH, have been detected in man and his environment. High levels of DDT and HCH have been reported in human blood, fat, and milk samples in India (Chatterjee et al. 1980). OCPs reported in the blood samples of human are listed in Table 10.7. Analysis of human exposure to selected OC compounds shows that the residue levels of *p, p′*-DDE and BHC were found to be persistent and higher in the human milk samples (Slorach and Vaz 1983), until the ban imposed on their use in the 1960s (ICMR 2001). High concentrations of both BHC and DDE were observed in the serum samples of the people who had direct exposure to the pesticides, namely agriculturalists and public health workers with few exceptions. The pesticide residue concentration in serum ranges from 0.006 to 0.130 ppm for BHC and from 0.002 to 0.033 ppm for DDE. This study reveals the presence of banned pesticides in human serum (Subramaniam and Solomon 2006).

Mathur et al. (2008) assessed the influence of OCPs upon the occurrence of reproductive tract cancers in women from Jaipur, India. Blood samples were collected from 150 women. In that group, 100 women suffered from reproductive tract cancers like cervical, uterine, vaginal, and ovarian cancers, while the rest did not suffer from cancers or any other major disease and were treated as control group. The pesticides detected were BHC and its isomers, dieldrin, heptachlor, and DDT and its metabolites. The data obtained indicate that the OCP residue levels were significantly higher in all the cancer patients as compared with the control group.

Pathak et al. (2008) analyzed the levels of OCP residues in maternal and cord blood samples of normal healthy women with full-term pregnancy to gain insight

Table 10.7 OCP residues (μg/L) in human blood reported in India

Location	α-HCH	β-HCH	γ-HCH	ΣHCH	*p, p*′-DDE	*p, p*′-DDD	*p, p*′-DDT	ΣDDT	**Source**
Lucknow	–	–	–	75	–	–	–	28	Kaphalia and Seth (1983)
Delhi	–	–	–	490	–	–	–	710	Ramachandra et al. (1984)
Delhi	–	–	–	–	–	–	–	301	Saxena et al. (1987)
Ahmedabad (rural)	–	–	–	148	37.2	1.33	8.83	47.7	Bhatnagar et al. (1992)
Ahmedabad	4.49	35.1	1.69	41.2	20.9	2.03	9.28	32.6[a]	Bhatnagar et al. (2004)
Punjab	–	–	–	57	–	–	–	65.2	Mathur et al. (2005)
Madurai	–	–	–	6.0–61	–	–	–	8.0–26	Subramaniam and Solomon (2006)
Bangalore (rural)	3.54	9.55	10.2	26.7[b]	5.67	3.01	3.81	10.6	Dhananjayan et al. (2012c)

[a] Total DDT (*p, p*′-DDE, *p, p*′-DDD, *p, p*′-DDT, *o, p*′-DDT) in serum
[b] ΣHCH (α-HCH, β-HCH, γ-HCH, δ-HCH)
– not available

into the current status of pesticide burden in newborns in North India. HCH contributed the maximum towards the total OC residues present in maternal and cord blood followed by endosulfan, *p, p*′-DDE, and *p, p*′-DDT being the least. This is also the first report indicating endosulfan levels in this population. These data indicate a transfer rate of 60–70 % of these pesticides from mothers to newborns, and this high rate of transfer of pesticides is of great concern as it may adversely affect the growth and development of newborn.

Dhananjayan et al. (2012c) described exposure level of OCPs among workers occupationally engaged in agriculture and sheep wool-associated jobs in rural neighborhood of Bangalore City, India. Thirty participants were interviewed and informed consent was obtained before blood sample collection. The maximum concentrations of OCP were detected in blood samples of agriculture workers rather than sheep wool workers. Among the metabolites of HCH and DDT, lindane (γ-HCH) and *p, p*′-DDE contributed the most to the total OCPs. There were no differences in pesticide residues found between sex and work groups.

10.3.10 Breast Milk

Bioaccumulation of OCs has been linked to effects such as endocrine disruption because of their capability of altering hormonal balance (Kelce et al. 1997; Beard et al. 2000). Furthermore, exposure to OCs has also been associated with an increased risk of breast and prostate cancer, endometriosis, cryptorchidism, and hypospadias (Birnbaum 1994; Hosie et al. 2000). Particularly, infants are extremely vulnerable to pre- and postnatal exposure to OCs, resulting in a wide range of adverse health effects including possible long-term impacts on intellectual function (Jacobson and Jacobson 1996; Eskenazi et al. 2006) and delayed effects on central nervous system functioning (Ribas-Fito et al. 2003; Beard 2006). Human breast milk was used as one of the best indicators of long-term exposure of OCs because it is easy to obtain, can be collected non-invasively, and indicates the contaminant levels in maternal fat (Tanabe and Subramanian 2006). In addition, breast milk monitoring provides a means of estimating intake of OCs by breast-fed infants. Previous studies reported the existence of high levels of certain OCs, particularly HCHs and DDTs in human breast milk from southern India (Tanabe et al. 1990; Subramanian et al. 2007; Kunisue et al. 2002; Minh et al. 2003; Tanabe and Kunisue 2007).

Blood and milk samples were collected from lactating women who were divided into four groups on the basis of different living standards, viz., residence area, dietary habits, working conditions, and addiction to tobacco. The level of total OCPs in blood ranged from 3.319 to 6.253 mg/L, while in milk samples it ranged from 3.209 to 4.608 mg/L. The results are in concurrence with the reports from other countries (Kumar et al. 2006). Aulakh et al. (2007) have reported the occurrence of DDT and HCH insecticide residues in human biopsy adipose tissues in Punjab, India.

Generally, levels of DDTs in India were lower than those observed previously, indicating that the concentration of DDTs has been declining in the Indian environment. For example, DDTs in human breast milk from New Delhi collected in 1989 by Nair and Pillai (1992) were 3,700 ng/g lipid wt. and have declined in 2006 by 1,500 ng/g lipid wt. (Devanathan et al. 2009); the DDT levels in Mumbai have declined from 8,000 ng/g lipid wt. in 1984 (Ramakrishnan et al. 1985) to 450 ng/g lipid wt. in 2006 (Devanathan et al. 2009), and in Kolkata there was a decline from 4,800 ng/g lipid wt. in 1984 (Ramakrishnan et al. 1985) to 1,100 ng/g lipid wt. in 2006 (Devanathan et al. 2009) (Table 10.8). However, increase in DDT concentrations was observed in Chennai from 760 ng/g lipid wt. in 1988 (Tanabe et al. 1990) to 1,200 ng/g lipid wt. in 2003 (Subramanian et al. 2007). Past and ongoing usage of DDT for controlling vector-borne diseases and/or continuing intake of contaminated foods may be a plausible reason for such an increasing trend. The general declining trends confirm the positive effects of governmental and voluntary restrictions and prohibitions on the usage of DDT and other measures taken to minimize the pollution. Although the results show declining trends, worldwide comparison indicates that India is still at the top of DDT contamination levels. In general, levels of DDTs in India resemble those of other developing countries like Indonesia (Sudaryanto et al. 2006), Malaysia (Sudaryanto et al. 2005), and Cambodia (Kunisue et al.

Table 10.8 Concentrations of OCs (ng/g lipid wt.) in human breast milk from major cities in India

Compound	New Delhi[a] (*n*=21)		Mumbai[a] (*n*=26)		Kolkata[a] (*n*=17)		Chennai[b] (*n*=12)	
	Range	Mean	Range	Mean	Range	Mean	Range	Mean
Lipid (%)	0.73–4.5	2.1	0.76–4.5	2.4	0.90–6.8	2.6	0.72–3.3	1.8
p, p′-DDE	68–10,000	1,200	39–1,300	380	110–2,300	920	640–2,800	1,100
p, p′-DDD	<0.026–230	24	1.2–25	7.3	3.0–31	15	2.2–17	8.0
p, p′-DDT	2.0–1,900	210	1.7–330	68	1.2–680	200	41–140	94
ΣDDTs	140–12,000	1,500	47–1,500	450	240–2,800	1,100	750–2,900	1,200
α-HCH	<0.21–22	4.6	0.59–19	4.7	3.2–34	9.1	2.5–15	7.9
β-HCH	4.2–1,600	240	6.1–1,200	210	61–1,900	680	1,700–8,700	4,500
γ-HCH	<0.20–1,700	82	<0.20–3.2	1.1	<0.20–7.6	2.0	0.30–10	1.9
ΣHCHs	6.3–1,800	340	12–1,200	220	74–1,900	670	1,700–8,700	4,500

n number of individuals

[a] Devanathan et al. (2009), [b] Subramanian et al. (2007)

2004b), but relatively lower than those of China (Kunisue et al. 2004a), Hong Kong (Wong et al. 2002), and Vietnam (Minh et al. 2004). The levels were one order of magnitude higher than the industrialized nations such as Japan (Kunisue et al. 2006), UK (Harris et al. 1999), Sweden (Nore'n and Meironyte 2000), Germany (Schoula et al. 1996), and Canada (Newsome and Ryan 1999). The elevated levels of DDTs in developing countries in the tropical region, including India, may be because considerable portions of DDT are still applied for malaria control and sanitary purposes. This contamination pattern implies that developing countries in the tropical region play a major role as a source of DDTs.

Among the DDTs, *p, p′*-DDE was the main compound found in many of the recent studies, accounting for more than 75% of the total DDTs concentrations, suggesting wide usage in the past and long-term accumulation of DDTs in humans. Due to various biological processes, metabolism of DDTs may occur throughout the food chain and, therefore, it can be suggested that the presence of *p, p′*-DDE in human tissues is derived not only from direct ingestion of *p, p′*-DDT but also from the ingestion of *p, p′*-DDE previously degraded in the environment. However, *p, p′*-DDT contributed to 15–20% of the total DDT concentrations in milk which also could be due to fresh exposure to DDT. The proportion of *p, p′*-DDT reported in the recent studies was relatively higher than other studies in developing countries such as Indonesia (Sudaryanto et al. 2006); 5–6% in China (Kunisue et al. 2004a); 8% in Phnom Penh, Cambodia (Kunisue et al. 2004b); and 8–12% in Vietnam (Minh et al. 2004). These results could be interpreted as an evidence for continuous fresh exposure to DDT in India, maybe from the malaria eradication program.

Although the usage of technical HCHs in India has been banned since 1997 for agriculture, the government still allows using HCHs for public health purposes and on certain crops (Li 1999). Furthermore, the government is also encouraging the replacement of technical HCHs with lindane (Gupta 2004). Relatively higher concentrations of HCHs were reported in human milk from Kolkata (670 ng/g lipid wt.) than in those from New Delhi and Mumbai (340 and 220 ng/g lipid wt., respectively) (Devanathan et al. 2009). These findings clearly show that the usage pattern of pesticides and exposure levels are different among the regions in India. As observed for DDTs, HCHs also show declining trends in New Delhi from 13,000 ng/g lipid wt. in 1997 (Banerjee et al. 1997) to 340 ng/g lipid wt. in 2006 (Devanathan et al. 2009), coinciding with the restrictions and prohibition on HCH usage in India. However, in Chennai, situated in the southern part of India, an increasing trend of HCH use was observed between 1988 (2,900 ng/g lipid wt; Tanabe et al. 1990) and 2003 (4,500 ng/g lipid wt.; Subramanian et al. 2007) which may be due to precarious use of HCHs and/or consumption of HCH-contaminated food.

Among HCH isomers, β-HCH was the predominant isomer contributing 80–96 % of the total HCH. β-HCH is the most persistent and bioaccumulative form and is eliminated slowly from the body. In addition, the ratio between different HCH isomers changes from lower tropic level in food chains to human milk, resulting in the more persistent β-HCH being predominant in human milk (Solomon and Weiss 2002). Interestingly, in all cities in India, α-HCH was higher in multipara mothers than in primipara, which also could be interpreted as an evidence for continuous intake of technical HCH. In general, γ-isomer in the present study accounted for lesser contribution to total HCHs. Generally, it was known that the levels of OCs in human breast milk were positively correlated with the age of mothers (Bates et al. 1994; Albers et al. 1996).

DDTs and HCHs were the most prevalent of OCs in human breast milk from India, HCHs being more predominant in southern part and DDT in other parts of India (northern, western, and eastern). Differences in accumulation patterns of OCs indicate region-specific usage of chemicals in India. Except in Chennai, levels of DDTs and HCHs in the present study were lower when compared to previous observations, indicating the effects of bans and restrictions imposed. No significant correlation was observed between concentrations of OCs in human breast milk and age of mothers and there was no difference in the concentrations between primipara and multipara mothers. The estimated infant daily intake of OCs shows that the intake of HCHs through lactation exceeded the TDI, which is of concern to infant health. Comprehensive studies on the OCs contamination in India are, therefore, necessary to understand the source and evaluation of possible long-term impacts of OCs (Devanathan et al. 2009). HCH isomers, endosulfan, malathion, chlorpyrifos, and methyl parathion were monitored in human milk samples from Bhopal, Madhya Pradesh. The endosulfan concentrations were highest and exceeded the HCH, chlorpyrifos, and malathion concentrations by 3.5-, 1.5-, and 8.4-fold, respectively. Through breast milk, infants consumed 8.6 times more endosulfan and 4.1 times more malathion than the average daily intake levels recommended by the WHO (Sanghi et al. 2003).

10.3.11 Biochemical Changes in Humans

Although 80 % of pesticides produced annually in the world are used in developed countries, less than half of all the pesticide-induced deaths occur in these countries. A higher proportion of pesticide poisonings and deaths occur in developing countries where there are inadequate occupational safety standards, ineffective protective clothing and washing facilities, insufficient enforcement, poor labeling of pesticides, illiteracy, and insufficient knowledge of pesticide hazards (Pimental and Greine 1996). Because farmers and farm workers directly handle 70–80% of the pesticides they use, they are at the greatest risk of exposure (McDuffie 1994). Most people do not realize that they are being poisoned by the pesticides, because many symptoms of pesticide poisoning are similar to other health problems, for example, skin rashes and dizziness. Apparently, therefore, a large number of acute pesticide poisonings each year go undiagnosed and unreported. Due to heavy pesticide exposure, various chronic effects such as brain and nervous system damage, cancer, birth defects, miscarriages, and still births have been reported. Few surveillance studies have been conducted in India on high-risk population groups involved in the spraying of pesticides in field conditions (Rupa et al. 1991; Gupta et al. 1995; Srivastava et al. 1995; Chaudhuri 2000).

The biochemical effects produced by certain pesticides can be enzyme induction or enzyme inhibition. The effect of pesticides may be detected by ensuring biochemical changes even before adverse clinical health effects can occur. Organophosphorus and carbamate pesticides are inhibitors of cholinesterase. Altered liver enzyme activities have been reported among pesticide workers exposed to organophosphorus pesticide alone or in combination with OC or other pesticides. More recently, various studies from several parts of the world revealed the toxic effects of pesticides on human beings especially by elucidating free radical mechanism, which can be confirmed by the direct measurement of lipid peroxidation by-products such as malondialdehyde (Ko et al. 1997). There has been no study so far on the biochemical aspects of environmental health with special reference to grape cultivation from developing countries.

Dhananjayan et al. (2012b) estimated the cholinesterase activity in blood samples of agricultural workers engaged in vegetables and grape cultivation. The results showed a marked inhibition in acetylcholinesterase (AChE) and butyrylcholinesterase (BChE) activity among agricultural workers (exposed subjects) compared to control subject. There was a statistically significant reduction in enzyme activity in both AChE (14%) and BChE (56%) among exposed groups (Patil et al. 2003). A total of 85 healthy male pesticide sprayers in grape garden exposed to different classes of pesticides for 3–10 years were compared with 75 controls matched for age with respect to serum cholinesterase, serum total protein, albumin, aspartate aminotransferase (AST), alanine transaminase (ALT), and hematological parameters such as Hb, Hct, red blood cell (RBC), and serum lipid peroxidation. Serum lipid peroxidation was estimated in the form of thiobarbituric acid reactive substances (TBARSs) produced. A significant decrease was observed in serum cholinesterase, serum total proteins, albumin, and hematological parameters, viz., Hb, Hct, and RBC. A significant increase in lipid peroxidation, AST, and ALT was observed in the exposed group when compared with control. These results suggest that the long-term exposure of various pesticides on sprayers of grape garden affects liver and heme biosynthesis and decreases serum cholinesterase.

Singh and Kaur (2012) presented a study that had been carried out to examine the acute symptoms of pesticide spraying in the farm workers of three villages in

Talwandi Sabo block of Bathinda district of Punjab, a cotton-growing area with high usage of pesticides. This is an exploratory health study recorded face-to-face with pretested questionnaire. A total of 108 male sprayers from villages of Bangi Nihal Singh (34), Jajjal (39), and Mahi Nangal (35) were field interviewed about the immediate impact of pesticides during spraying season from September to October 2003. Majority of the sprayers complained of having nausea, itchiness of the eyes, pain while urinating, discolored nails, nails dropping off, swollen fingers, sleeplessness, headache, excessive sweating, and skin rashes. Immediate attention should be given to the implementation of proper awareness programs for pesticide workers. Also, practices like integrated pest management, organic farming, biopesticides, and crop diversification should be promoted.

The analysis of OCPs using confirmatory techniques, e.g., GC–MS or GC–MS/MS, gives unambiguous results and is, therefore, extremely important for such monitoring studies as has been reviewed in the chapter (Selvi et al. 2012; Maurya et al. 2013).

10.4 Conclusion and Recommendation

OC insecticides are still used for agriculture and public health purposes raising a major concern on their residual concentrations in the environment. In developing countries, OCs are preferred since they are cheap and more effective. Nevertheless, only a minor fraction of applied OCPs reaches the target species. The excess pesticide moves through the environment, potentially contaminating soil, water, and all other biotic matrices. In India, agricultural fields are generally located in plains, highlands, and valleys; rivers and streams carry pesticide residues into estuaries and into the ocean contaminating the sediments which act as a sink to most OC residues in aquatic environments. OCP residues detected in foodstuffs, fishes, birds, and humans reflect the usage of various types of OCs in this country. Although *p, p′*-DDT had been banned in 1997 following the Stockholm Convention on POPs, the presence of DDT in environmental matrices was reported in many studies. In several cases, the DDT levels exceeded the recommended levels in comparison with the sediment quality guidelines and could, thus, cause acute biological impairments. Contributions of DDT metabolites vary in different Indian regions predominated by *p, p′*-DDT and *p, p′*-DDD. HCH and DDT residues in fish in India were lower than those in the temperate countries indicating a lower accumulation in tropical fish, which might be related to rapid volatilization of this insecticide in the tropical environment.

This review of data on the environmental fate and effects of OCPs in India suggests a causal relationship between the application of pesticides for public health purposes and agriculture use. The comparison of concentrations reported in various research studies with reference values from various national and international standard documents leads to the assumption that the presence of these OCPs has created serious detrimental effects on fish, wildlife, humans, and the environment.

However, the long-term effects of pesticide were reported for various biological systems. Most of the data suggest that DDTs and HCHs were the most prevalent OCs within different compartments of the ecosystem, HCHs being more predominant in the southern part and DDT in other parts of India. Because of their high biological activity and, in some cases, of their persistence in the environment, the use of pesticides may cause undesired effects on human health and the environment. In view of their potential toxic and persistent nature, there is a pressing need for their control and monitoring in the environment. As far as wildlife refuges are concerned, the literature is negligible. In India, unfortunately, no historical data on the levels of OCPs are available in many species of wildlife. It is needless to say that each and every organism in the ecosystem will get affected by persistent OCPs.

At present, POPs are banned in most of the countries. Trade in these substances is also restricted. However, some stock of these compounds is still available in the market. Some developing countries allow some use of these compounds for public health purposes, while their use in developed countries for any purpose is banned. Rotation of pesticides used and integrated pest management practices have been recommended as possible solutions to this problem. The level of education in developing countries is low. The challenge for the future is to generate resources for education and research. Disposal of pesticides requires heavy investment that is beyond the financial resources available to most developing countries. The challenge here is first to develop human capacity in waste management and research in order to install facilities for POP disposal. The need of the hour, therefore, is regular monitoring attempts at pesticide residue evaluation in order to give a baseline for further studies in these disciplines. Hence, comprehensive studies on the OCs contamination in India are, therefore, necessary to understand the source and evaluation of possible long-term impacts of OCs.

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