

Organochlorine pesticide residue status in dry fish of Bangladesh and their risk assessment: a review

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Abstract

Organochlorine pesticides including DDT and its derivatives used in dry fish in Bangladesh till 2019 has been reviewed in this article. Level of organochlorine pesticide residues remain in dry fish in Bangladesh were collected from previously published peer-reviewed journals and the risk assessments of those pesticides were also addressed in this review article. In this article, level of residues of DDT, aldrin, dieldrin, endrin, endrin ketone, heptachlor, heptachlor epoxide has been reported for several most popular marine dry fish: Ribbon fish (*Lepturacanthus savala*), Chinese pomfret (*Pampus chinensis*), Bombay duck (*Harpodon nehereus*), Sin Croaker (*Johnius dussumieri*), *Shrimp sp.* (crustaceans) and fresh water dry fish: Chepa Sutki (*Puntius sophore*) and several other dry fishes namely Nona Ilish (*Hilsa ilisha*), Boro Chingri (*Macrobrachium rosenbergii*), Gura Chingri (*Leander styliferus*), Kaski (*Corica soborna*), Chapila (*Gudusia chapra*), Shoal (*Channa striatus*), Deshi Chanda (*Chanda ranga*), Ghonia (*Labeo gonius*). Intake of significant amount of these pesticides is a matter of health concern. These risks should be re-assessed periodically to build awareness of harmful effect of these insecticides on dry fish. Government of Bangladesh should take all the necessary steps to prevent the use of these toxic pesticides in dry fish processing and preservation.

Keywords: Dry fish; Organochlorine pesticide residue; Health hazard.

I. INTRODUCTION

In Bangladesh, fish plays a central role in dietary patterns, livelihoods and culture being a fish-loving nation which means “Bengali by fish and rice”. Fish is by far the most commonly consumed animal-source food across all population groups, at an average of 62.58 grams per day against the daily requirement of 60 grams (19.71 kg/person/year) [1]. Fish is an important diet staple, providing a rich source of micro-nutrients and accounting for 60% of animal protein intake [2]. Dry fish (SUTKI in Bengali) is low cost dietary protein source and every year a significant portion of fresh fish is sun dried due to high market demand and a substitute of scarcity of fresh fish. Besides protein source dried fishes are also rich in vitamins and minerals, which are often overlooked in developing countries [3, 4, 5, 6]. About 15% of fishes are cured for mass people consumption at the scarcity of fresh fishes in Bangladesh [7]. Dry fish consumption frequency is very satisfactory in the Southeast Asian countries where people in Bangladesh consume dry fish at least once a week in their meal [8]. Special flavor is highly relished by different ethnic people. The product of dried fish is easily transportable, marketable and storable [21]. Research shows that increasing fish consumption and dietary diversity can affect significant developmental changes. The culture and consumption of fish therefore has important implications for national food and nutrition security, poverty and growth [10]. The prime source of high-quality protein is fish, which provides 14–16% of the animal protein consumed worldwide [11]. Over one billion people across the world consume fish as their primary source of animal protein [12]. Thus fish either harvested from natural source(s) or cultured artificially and the fish products have great importance as human food worldwide. Both capture fisheries and aquaculture play significant roles in fish production and consumption in Bangladesh. The country was ranked third in producing fish from inland water-bodies, behind China and India, according to Food and Agriculture Organization (FAO) report “The State of World Fisheries and Aquaculture 2018”, Bangladesh ranked 3rd in inland open water capture production and 5th in world aquaculture production. Currently Bangladesh ranks 4th in tilapia production in the world and 3rd in Asia [1].

Unfortunately, there are recurrent concerns regarding traditionally dried fish quality sanitation and hygiene, as is seldom practiced. The customary drying of fish is primarily performed by poor and ignorant makers whose action comprises of spreading fish on split bamboo mats, solid floors, or raised stages or draping fish over bamboo posts and bars [13]. One of the serious issues related with dried fish is infestation by varieties of insects such as mites, beetles, and blowflies [7, 14, 15]. The extent of damage done by larvae and adult insects (loss of soft tissue and weight loss) largely depends on the drying speed, the size of the fish, and whether the fish is salted [16]. In Bangladesh, producers often use organochlorine pesticides such as dichlorodiphenyltrichloroethane (DDT) powder (locally known as white powder), dichlorvos, heptachlor, or whatever they get within their reach to protect dried fish from infestation

and increase storage time [17- 20]. Several analyzes indicate troubling toxins in fish such as DDT and heptachlor in Bangladesh [22]. In Kuakata (a fish processing zone in Bangladesh), high level of DDT powder is used though Bangladesh banned the 'dirty dozen' in 1997 [23] and there is no statistical figure about these organochlorine insecticides in Bangladesh [24]. Such pesticides have led to the development of resistance to pests [25], resulting in higher doses of pesticides [24] being applied.

In Bangladesh, however, organochlorine pesticides were banned in 1993 [26] due to their high human and environmental toxicity, acute persistence, bioaccumulation potential and biomagnification in the food chain [27, 28]. Among different gatherings of pesticides, the utilization of organophosphorus insecticides has turned out to be progressively mainstream in Bangladesh. Organochlorine insecticides are the synthetic organic insecticides that contain carbon, chlorine and hydrogen. They are profoundly diligent in the life form and environment because of their low water dissolvability and high lipophilicity [9]. Because of their tirelessness and modest value, the OC insecticides such as dichlorodiphenyltrichloroethane (DDT), aldrin, endosulfan had been generally utilized for pest control [9]. In any case, it was later found that OC insecticide residues were gradually discharged into aquatic and earthbound biological systems [9]. These residues can be moved and biomagnified through food chain. Along these lines critical degrees of OC insecticides could be amassed and caused unfavorable wellbeing impacts in creatures and higher trophic levels, including human [9, 29]. OC insecticides contamination in animal tissues was linked to adverse effects on reproductive system such as reduced penis size of American alligator *Alligator mississippiensis* in Lake Apopka, Florida, USA, and abnormality in reproductive functions of Florida red-belly turtles *Chrysemys nelsoni* [30]. Since human populace might be correspondingly in danger from these synthetic chemicals. The OC insecticides have been banned in numerous nations (1970 in Sweden; 1971 in Japan; 1972 in USA). India banned DDT for agricultural purpose in 1989 however keeps on utilizing between 5,000-10,000 kg/year for malaria control. In India, the indiscriminate and injudicious use of OC insecticides have led to the contamination of water bodies near to lands, rivers and estuarine bodies [31, 32] resulting in high concentration in aquatic life especially fish, prawns, otters, shrimps etc. [33, 31]. The consumption of biota from contaminated aquatic body is considered to be an important route of exposure to OC insecticides [34] in humans resulting in variety of health hazards particularly endocrine dysfunction, births defects, breast cancer, lower sperm count and testicular cancer [35, 36, 37]. OC insecticides are still detectable in fish from various waterways [38, 39]. Use of organochlorine pesticides (OCP) including DDT have been banned in Bangladesh after signing Stockholm Convention [40, 41] but evidences of the presence of OCP were found in environmental samples like fish, dry fish, and poultry feed [87, 43] and also through the food chain in human blood samples [44, 45]. The cause may be illegal trafficking of banned OCP from neighboring countries where OCP are allowed to use in health sector and pilferage from large stockpiles of OCP in the three godowns of Bangladesh [42]. Fish is one the most suitable bio-concentrators to identify OCP [46].

Social concerns about pesticides in fish are due to their presence in muscle tissue of edible species. Recent reports state the role of fish in human POPs exposure through dietary consumption of freshwater [47, 48, 49] and marine species [50]. Risk assessment based on acceptable daily intake (ADI) criteria of POPs findings have been discussed [51, 47, 52, 53, 48, 54, 55]. From a regulatory point of view, pesticide occurring in fish cannot be related to classical MRLs (maximum residue limit) based on Good Agricultural Practices of production. Pesticide MRLs in fish should be classified as extraneous MRLs (E-MRLs) as pesticides are true water contaminants that are incorporated in fish tissues. Nevertheless, although advising to analyze only the edible portion of fish, the Codex Alimentarius currently rules no E-MRL for freshwater fish for the 29 fish species listed [56]. On the other hand, in marine fish only lindane occurring at 0.01 mg/kg is listed. No MRL for pesticides has been ruled for trouts, eels, salmons, sardines, herrings, bonito, tuna and other 61 different widely consumed fishes all over the world at Codex Alimentarius [56]. This unregulated scenario implies that pesticide residues should not be detected in edible fish samples. Some countries such as Russian Federation and Canada, settled particular MRLs for DDT and lindane at 2 mg/kg. In the European Union, if the precautionary principle is applied, pesticides should not be in food at levels higher than 0.01 mg/kg. Considering the increasing amount of evidence of pesticide occurrence in edible fish tissues, food safety issues should be urgently addressed by regulatory agencies. The MRLs settling for eventual pesticides either CUPs or POPs in fish is related exclusively to toxicological parameters, where the acceptable daily intake (ADI) should be the ruling parameter. Within this frame, the global contribution of fish intake to any national diet has to be considered.

II. OVERALL AIMS OF THIS STUDY

This study was performed for the accumulation of fundamental data on Organochlorine pesticides including DDT and its metabolites residues remain in dry fishes in Bangladesh as pesticides are used to preserve dry fishes in Bangladesh. The data were collected from the published peer-reviewed journals. All published data will be compared and try to find the present status of DDT, aldrin, dieldrin, endrin, endrin ketone, heptachlor, heptachlor epoxide residues in dry fish. Organochlorine pesticides (OCP) including DDT have been banned in Bangladesh after signing Stockholm Convention [40, 41] but evidences of the presence of OCP are still detectable from collected samples of dry fish. Due to lack of awareness, the farmers are using these pesticides in dry fish to extend shelf life. Constant monitoring is required to detect it and build awareness of its severe effect on nature and human.

III. Materials and Methods

A. Data analyzed and study area

Reviewed papers were selected based on banned organochlorine insecticides used in dry fish preservation in Bangladesh (Table 2). OCPs were selected to represent each main use type DDT, aldrin, dieldrin, endrin, endrin ketone, heptachlor, heptachlor epoxide. The chemical structures of the selected OCPs are shown in Fig. 1, and the modes of action and classifications of the OCPs are shown in Table 1. Data for the risk assessments of the OCPs were obtained from previous publications [57,58] and from Table 2.

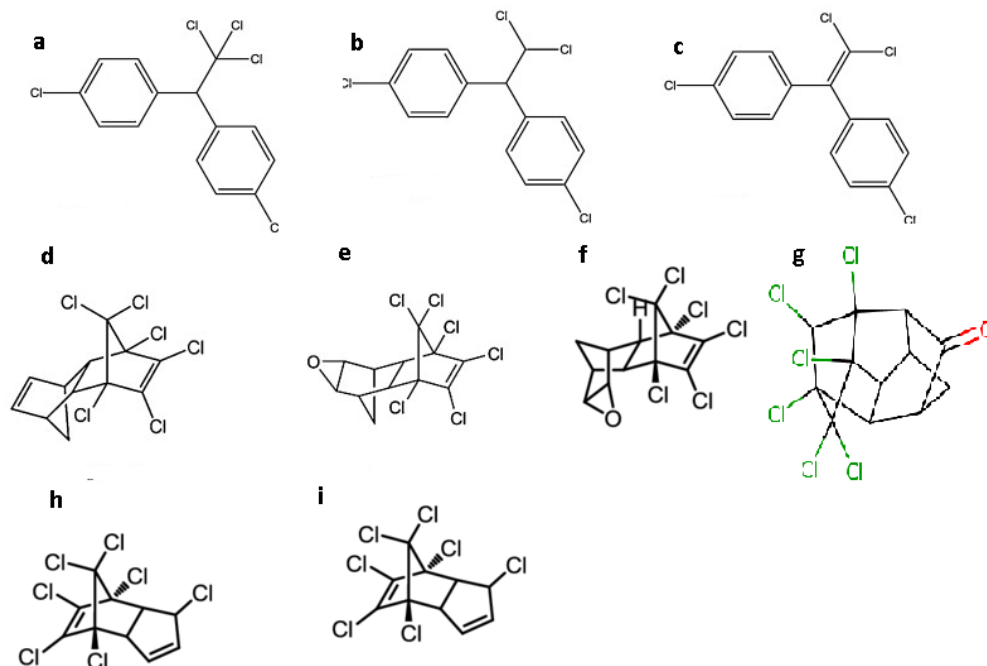


Fig. 1: Chemical structures of reviewed organochlorine pesticides: DDT (a); DDD (b); DDE (c); aldrin (d); dieldrin (e); endrin (f); endrin ketone (g); Heptachlor (h); Heptachlor epoxide (i).

Table 1: Names, classes, and modes of action of the pesticides that were assessed ^a

Compound	Chemical class	Mode of action
DDT	Organochlorine Insecticide	Non-systemic stomach and contact action. Sodium channel modulator.
DDD	Organochlorine Insecticide, Metabolite	Non-systemic stomach and contact action
DDE	Organochlorine Insecticide, Metabolite	Central nervous system stimulant. GABA-gated chloride channel antagonist. A weak androgen receptor antagonist.
Aldrin	Organochlorine Insecticide	Central nervous system stimulant. GABA-gated chloride channel antagonist. Also stomach and contact toxin
Dieldrin	Organochlorine Insecticide	Central nervous system stimulant. GABA-gated chloride channel antagonist. Contact and stomach action
Endrin	Organochlorine Insecticide, Avicide, Rodenticide	Broad-spectrum, with contact and stomach action. Is a chloride channel-blocking agent.
Endrin ketone	Organochlorine Insecticide,	Endrin antagonizes the action of the neurotransmitter gamma-amino butyric acid (GABA) acting at the GABA-A receptors, effectively blocking the GABA-induced uptake of chloride ions and causing hyperexcitability of the central nervous system.
Heptachlor	Organochlorine Insecticide	Persistent, non-systemic contact and stomach poison with some fumigant action. Is a chloride channel-blocking agent.
Heptachlor epoxide	Organochlorine Insecticide	Heptachlor epoxide is a central nervous system stimulant. It non-competitively blocks neurotransmitter action at gamma-amino butyric acid receptors, resulting in overstimulation of the nervous system. Heptachlor epoxide is also believed to exert carcinogenic effects by activating key kinases in signalling pathways and inhibiting

		apoptosis
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^a. Source of all OCPs pesticides data in this table obtained from PPBD, 2018[58]. And <http://www.t3db.ca>

Table 2: List of pesticide residues in Dry Fish found by several researchers in Bangladesh

Matrices	Area of collection	No. of sample analyzed	Detected pesticides	Percentage of contaminated samples	Percentage of samples above MRL	References
Chepa shutki (dry fish)	Dhaka City, Gazipur	10	DDT, Aldrin, Dieldrin, Endrin, Heptachlor	100	100	[59]
Loitta, Kanchki, Mola, Paysha, Chapa, Churi, Chingri, Hangor, Chanda, Shidhol	Bogura, Chattogram, Khagrachori, Mymensingh, Rajshahi, Rangpur, Dinajpur and Dhaka	60	Endrin, Heptachlor epoxide, Endrin ketone	33	15	[60]
Ribbon fish (<i>Lepturacanthus</i>), Chinese pomfret (<i>Pampus chinensis</i>), Bombay duck (<i>Harpodon nehereus</i>) and Shrimp sp.	Khulna, Chattogram and Cox's Bazar	36	DDT	100	100	[61]
Ribbon fish (<i>Lepturacanthus savala</i>), Sin Croaker (<i>Johnius dussumieri</i>), Bombay duck (<i>Harpodon nehereus</i>) and Shrimp (<i>mixed species</i>)	Chittgong, Cox's Bazar	24	DDT	100	16	[17]
Dry Puti (<i>Puntius sophore</i>), Chepa Sutki (<i>Puntius sophore</i>), Nona Ilish (<i>Hilsa ilisha</i>), Boro Chingri (<i>Macrobrachium rosenbergii</i>), Gura Chingri (<i>Leander styliferus</i>), Kaski (<i>Corica soborna</i>), Chapila (<i>Gudusia chapra</i>), Shol (<i>Channa striatus</i>), Deshi Chanda (<i>Chanda ranga</i>), Ghonia (<i>Labeo gonius</i>)	Dhaka City	50	DDT DDE DDD	100	2	[41]
Bombay duck (Loitty), Ribbon fish (Chhuri), Anchovy (Teilla phasa), Croaker (Poa), Chinese pomfret (Rupchanda) and Indian salmon (Lakhua).	Dhaka City	6	Heptachlor DDT	100	33.33	[62]
Bombay duck (Loitty), Ribbon fish (Chhuri), Shrimp (Chingri), Chinese pomfret (Rupchanda) and Indian salmon (Lakhua).	Chattogram	20	Heptachlor DDT	100	None	[20]
Bombay duck (Loitty), Ribbon fish (Chhuri), Shrimp (Chingri)	Dhaka, Chattogram, Khagrachori	12	DDT, Heptachlor	100	None	[63]

B. Risk assessment for OCPs

I.B. Human health risk estimation

The human health risk was evaluated based on the concentration of pesticides residues in Dry fish. The estimated daily intake (EDI) was calculated based on the residue concentrations measured in the different food products [64]. This was performed using Equation (1)

$$EDI_i = \frac{C_i \times IR}{BW} \quad (1)$$

Where EDI_i is the estimated daily intake of ith pesticide, C_i is the mean residual concentration of ith pesticide (mg kg⁻¹), IR is the estimated daily ingestion rate (kg d⁻¹) and BW is the hypothetical average body weight (kg) set at 10 kg for children and 60 kg for adults [65-67]. A daily ingestion rate of 0.00274 kg d⁻¹ dry fish per person was used based on local food consumption data [68]. OCP tissue concentration data for dry fish previously obtained from Table 2, were incorporated into the risk assessment.

II.B. Non-carcinogenic risk

To assess the non-carcinogenic risk posed by the consumption of contaminated fish to humans, a hazard quotient (HQ) approach was used [69]. HQ values were calculated for concentrations using Equation (2) as follow:

$$HQ_i = \frac{EDI_i}{ADI_i} \quad (2)$$

ADI is the acceptable (tolerable) daily intake [70, 71] assumed to be a safe concentration for lifetime exposure. For the purposes of a preliminary quantitative risk assessment, a $HQ \leq 0.2$ is considered to indicate negligible adverse health effects, while HQ values exceeding this threshold require a further detailed risk assessment or risk management measures to be undertaken [72]. Subsequently, cumulative hazard indices (HIs) were calculated as the sum of all individual HQs at their respective concentration. HI values > 1 indicate potential adverse health effects requiring a detailed human health risk assessment, while values < 1 indicate no potential adverse health implications.

III.C. Cancer risk

In assessing the potential carcinogenic risks posed by OCPs through the consumption of contaminated dry fish, both cumulative lifetime cancer risk (LCR) estimates and hazard ratios (HR) were derived based on US EPA guidelines [66]. The LCR was calculated by multiplying the EDI by the cancer slope factor (CSF). LCR is defined here as the increased probability of developing cancer over a lifetime due to continuous exposure to a carcinogen. According to the US EPA, a risk level below one in a million (10^{-6}) is considered acceptable, between one in ten thousand and one in a million (10^{-4} and 10^{-6} , respectively) implies a potential risk and greater than one in ten thousand (10^{-4}) is considered a high cancer risk [66]. HR for carcinogenic effects were calculated following [73], Equation (3):

$$HR = \frac{EDI_i}{BMC} \quad (3)$$

where BMC is the benchmark concentration. The BMC for cancer effect is derived from the US EPA CSF obtained from the US EPA Integrated Risk Information System (IRIS). The BMC was calculated using the expression:

$$BMC = \frac{\text{Risk} \times BW}{IR \times CFS}$$

Where the cancer risk is set to one in one million (10^{-6}) and IR is the amount of a particular food product consumed per kg body weight of individual per day. An $HR > 1$ implies that there is a potential human health risk from the consumption of contaminated food [74].

The present study is based on screening a number of literatures that documented some of the impact of the use of DDT and its metabolites in Bangladesh (Table 2).

IV. Results and discussion

IV.A. Trends in use of selected OCPs in Dry fishes of Bangladesh

The organochlorine pesticides namely DDT, aldrin, dieldrin, endrin, endrin ketone, heptachlor, heptachlor epoxide were found in different dry fishes by several researchers. The present study have been undertaken in order to provide the preliminary information on the concentration of DDT and heptachlor found in marine dry fish and the concentration of DDT, aldrin, dieldrin, endrin, endrin ketone, heptachlor, heptachlor epoxide were found in fresh water dry fish.

IV.B. Marine Dry Fish:

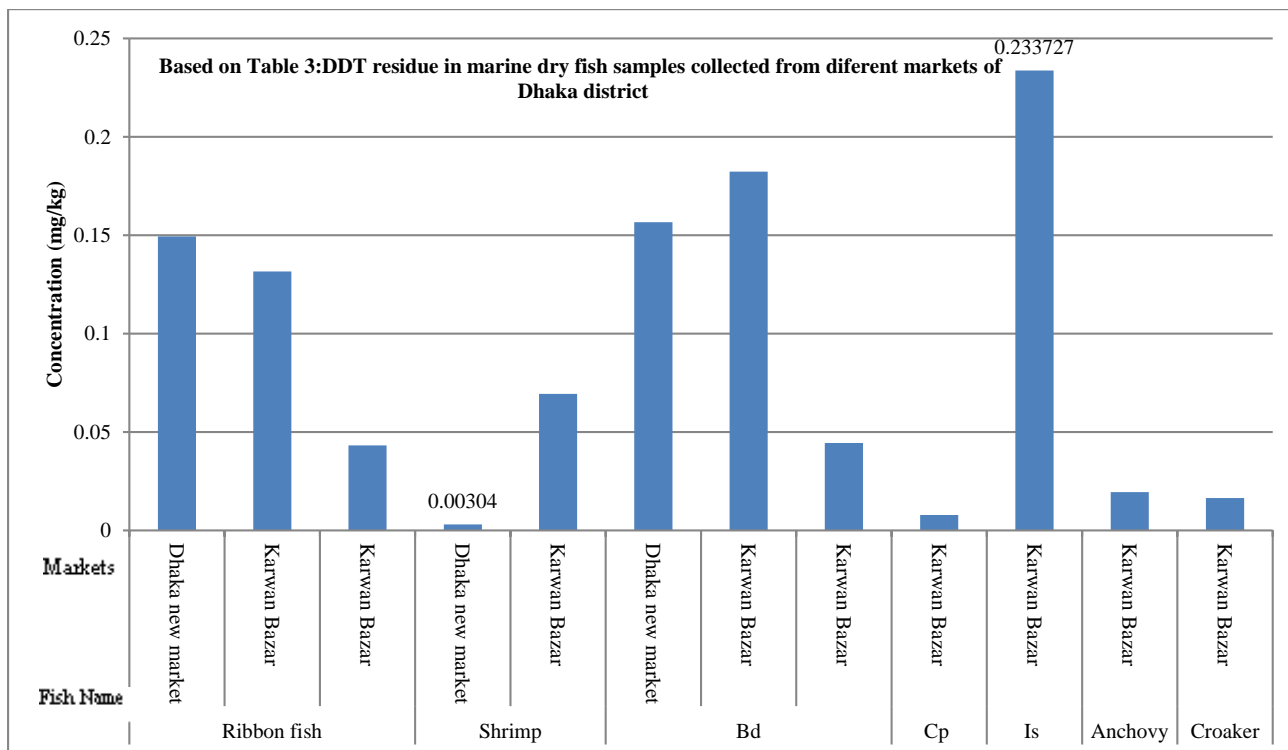
The detected DDT residue found in the collected marine dry fish samples from different markets of Dhaka district are summarized in Table 3. Where ribbon fish samples collected from Dhaka new market and Karwan Bazar, shrimp from Karwan Bazar, Bombay duck from Dhaka new market and Karwan Bazar, indian salmon from Karwan Bazar markets exceeded the MRL set by European Commission [75].

Table 3: DDT pesticide residue (mg kg^{-1}) in the marine dry fish samples collected from different markets of Dhaka district.

Name of Dry Fish	Area of Collection	Level of residue (mg/kg)	MRL (mg/kg)	References
Ribbon fish	Dhaka new market	0.1494	0.05	[63]
	Karwan Bazar	0.1316		[63]
	Karwan Bazar	0.043158		[63]
Shrimp	Dhaka new market	0.00304		[63]
	Karwan Bazar	0.0694		[63]
	Dhaka new market	0.1566	[63]	

Bombay duck	Karwan Bazar	0.1823	[63]
	Karwan Bazar	0.044395	[62]
Chinese pomfret	Karwan Bazar	0.007849	[62]
Indian salmon	Karwan Bazar	0.233727	[62]
Anchovy	Karwan Bazar	0.019416	[62]
Croaker	Karwan Bazar	0.016404	[62]

The level of DDT residues in all samples ranged from 0.00304 mg kg⁻¹ to 0.233727 mg kg⁻¹. Among the samples, the DDT was found comparatively lowest in shrimp and highest in indian salmon (Fig. 2).



*Bd = Bombay duck; Cp = Chinese pomfret; Sh=Shrimp; Is = Indian salmon.

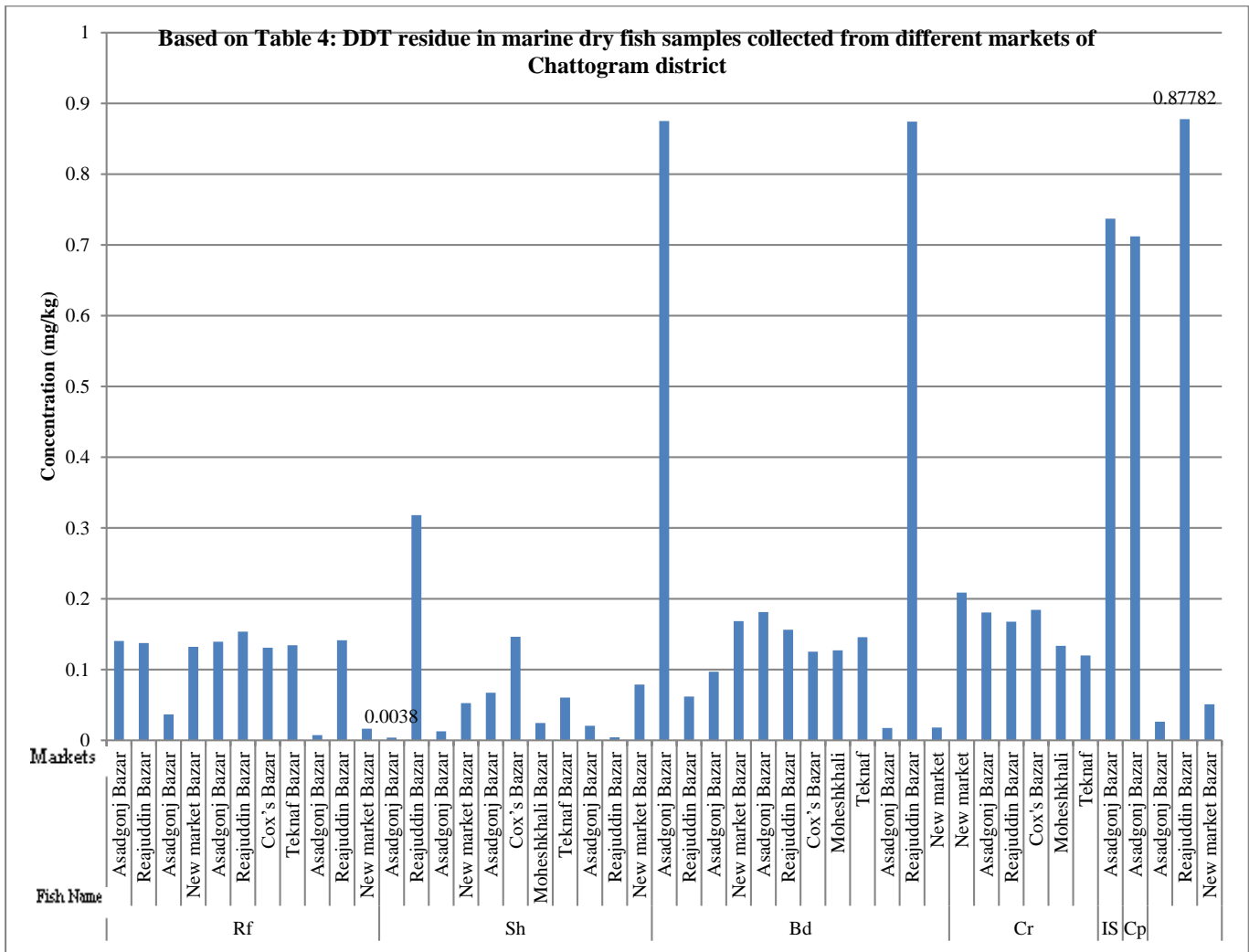
Figure 2: Level of DDT (mg kg⁻¹) in the marine dry fish samples collected from different markets of Dhaka district.

Table 4: DDT pesticide residue (mg kg⁻¹) in the marine dry fish samples collected from different markets of Chattogram district.

Name of Dry Fish	Area of Collection	Level of residue (mg/kg)	MRL (mg/kg)	References
Ribbon fish	Asadgonj Bazar	0.1404	0.05	[63]
	Reajuddin Bazar	0.1375		[63]
	Asadgonj Bazar	0.0366		[20]
	New market Bazar	0.13211		[17]
	Asadgonj Bazar	0.13934		[17]
	Reajuddin Bazar	0.15347		[17]
	Cox's Bazar	0.13085		[17]
	Teknaf Bazar	0.13428		[17]
	Asadgonj Bazar	0.00719		[61]
	Reajuddin Bazar	0.14143		[61]
	New market Bazar	0.01628		[61]

Shrimp	Asadgonj Bazar	0.0038	[63]
	Reajuddin Bazar	0.3182	[63]
	Asadgonj Bazar	0.0126	[20]
	New market Bazar	0.05244	[17]
	Asadgonj Bazar	0.0671	[17]
	Cox's Bazar	0.14637	[17]
	Moheshkhali Bazar	0.02437	[17]
	Teknaf Bazar	0.0604	[17]
	Asadgonj Bazar	0.02048	[61]
	Reajuddin Bazar	0.00427	[61]
	New market Bazar	0.07878	[61]
	Chinese pomfret	Asadgonj Bazar	0.7122
Asadgonj Bazar		0.02627	[61]
Reajuddin Bazar		0.87782	[61]
New market Bazar		0.05095	[61]
Indian salmon	Asadgonj Bazar	0.7372	[20]
Bombay duck	Asadgonj Bazar	0.875	[63]
	Reajuddin Bazar	0.619	[63]
	Asadgonj Bazar	0.0970	[20]
	New market Bazar	0.16836	[17]
	Asadgonj Bazar	0.18114	[17]
	Reajuddin Bazar	0.15617	[17]
	Cox's Bazar	0.12521	[17]
	Moheshkhali	0.12706	[17]
	Teknaf	0.14575	[17]
	Asadgonj Bazar	0.01742	[61]
	Reajuddin Bazar	0.87435	[61]
	New market	0.01799	[61]
Croaker	New market	0.20865	[17]
	Asadgonj Bazar	0.18063	[17]
	Reajuddin Bazar	0.16752	[17]
	Cox's Bazar	0.18422	[17]
	Moheshkhali	0.13352	[17]
	Teknaf	0.11986	[17]

The detected DDT residues were found in the collected marine dry fish samples from different markets of Chattogram are summarized in Table 4. It is observed that almost all samples were contaminated with DDT above the EU-MRLs [75], only shrimp samples may be differentiated. The concentration level of DDT in all marine dry fish samples collected from Chattogram ranged from 0.0038 mg kg⁻¹ to 0.87782 mg kg⁻¹. Among the samples, the DDT was found highest in chinese pomfret (0.87782 mg kg⁻¹) collected from Reajuddin Bazar and lowest in shrimp (0.0038 mg kg⁻¹) collected from Asadgonj Bazar (Fig. 3).



* Rf=Ribbon fish; Sh=Shrimp; Bd = Bombay duck; Cr = Croaker ; Is=Indian salmon ; Cp = Chinese pomfret.

Figure 3: Level of DDT (mg kg⁻¹) in the marine dry fish samples collected from different markets of Chattogram district.

Table 5: DDT pesticide residue (mg kg⁻¹) in the marine dry fish samples collected from different markets of Khulna district.

Name of Dry Fish	Area of Collection	Level of residue (mg/kg)	MRL (mg/kg)	References
Bombay duck	Khalishpur	0.0137	0.05	[61]
	Khuruskhul	0.01409		[61]
	Borobazar	0.26189		[61]
Chinese pomfret	New Market Bazar	0.00281		[61]
	Moilapota	0.05801		[61]
	Khalishpur	0.25047		[61]
	Nagirartek	0.03827		[61]
	Khuruskhul	0.04517		[61]
	Borobazar	0.05832		[61]
	Ribbon fish	Moilapota		0.25368
	Khalishpur	0.00373	[61]	

	Nagirartek	0.02062	[61]
	Khuruskhul	0.05824	[61]
	Borobazar	0.20913	[61]
Shrimp	Moilapota	0.58597	[61]
	Khalishpur	0.02139	[61]
	Nagirartek	0.01542	[61]
	Khuruskhul	0.05616	[61]
	Borobazar	0.16167	[61]

The detected DDT residues were found in the marine dry fish samples collected from different markets of Khulna district are summarized in Table 5. Where bombay duck collected from Borobazar, chinese promfret collected from Khalishpur, ribbon fish collected from Moilapota, Borobazar and shrimp collected from Moilapota, Borobazar markets exceeded the EU-MRLs[75]. The level of DDT residues in all collected marine dry fish samples from Khulna district were found ranged from 0.00281 mg kg⁻¹ to 0.58597 mg kg⁻¹. This study indicates highest concentration of DDT in shrimp (0.58597 mg kg⁻¹) collected form Moilapota market and lowest in chinese promfret (0.00281 mg kg⁻¹) collected from Khulna new market (Fig. 4).

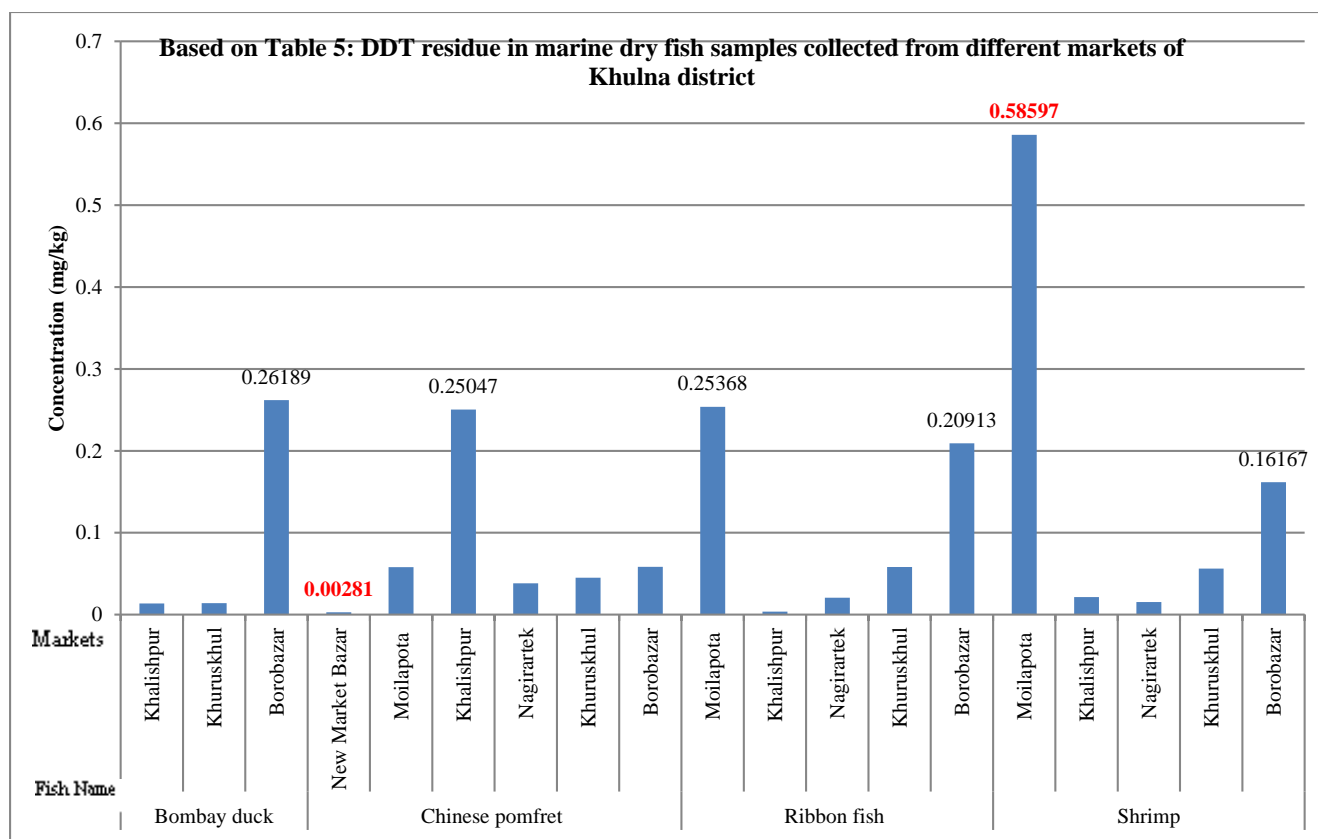


Figure 4: Level of DDT (mg kg⁻¹) in the marine dry fish samples collected from different markets of Khulna district.

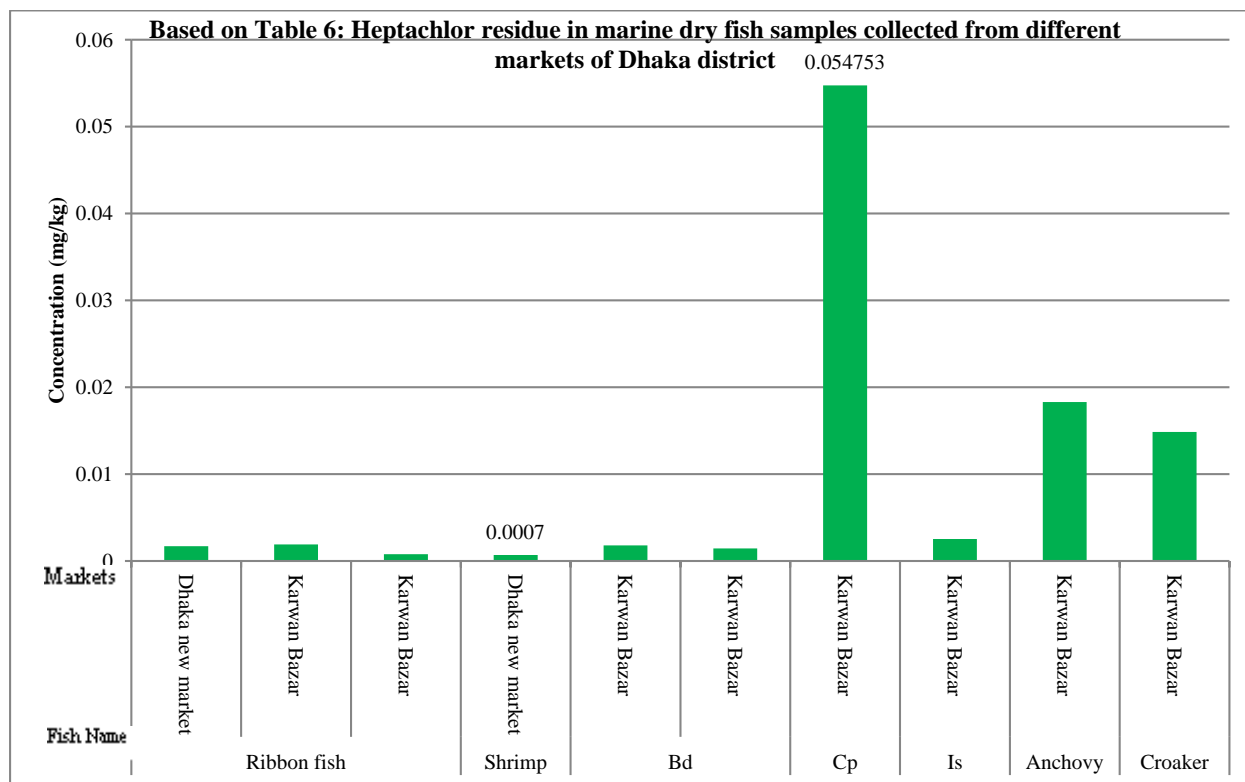
The ribbon fish samples of all places contained almost about same concentration of DDT. The concentration of DDT in shrimp including Dhaka, Chattogram and Khulna were ranged between 0.00304 mg kg⁻¹ to 0.5897 mg kg⁻¹. However, it is found that the concentration of DDT is depends on sample size, bigger size of shrimp was found to contained higher amount of DDT as the fishers pay much more attention during preservation of bigger size shrimp and use higher amount of DDT than the small size.

The concentration of another organochlorine pesticide, heptachlor, in the marine dry fish samples collected from different markets of Dhaka district are summarized in Table 6. Where heptachlor residue in chinese pomfret (0.054753 mg kg⁻¹) and anchovy (0.018306 mg kg⁻¹) collected from Karwan Bazar exceeded the EU-MRLs [75].

Table 6: Heptachlor residue (mg kg⁻¹) in the marine dry fish samples collected from different markets of Dhaka district.

Name of Dry fish	Area of Collection	Level of Residue (mg/kg)	MRL (mg/kg)	References
Ribbon fish	Dhaka new market	0.0017	0.01	[63]
	Karwan Bazar	0.0019		[63]
	Karwan Bazar	0.000778		[62]
Shrimp	Dhaka new market	0.0007		[63]
Bombay duck	Karwan Bazar	0.0018		[63]
	Karwan Bazar	0.001452		[62]
Chinese pomfret	Karwan Bazar	0.054753		[62]
Indian salmon	Karwan Bazar	0.002532		[62]
Anchovy	Karwan Bazar	0.018306		[62]
Croaker	Karwan Bazar	0.014856		[62]

It is observed that the range of heptachlor concentration in all samples were 0.0007 mg kg⁻¹ to 0.054753 mg kg⁻¹ (Fig. 5). The highest concentration of heptachlor found in chinese pomfret (0.054753 mg kg⁻¹) collected from Karwan Bazar and lowest in shrimp (0.0007 mg kg⁻¹) collected from Dhaka new market.



*Bd = Bombay duck; Cp = Chinese pomfret; Is = Indian salmon.

Figure 5. Level of Heptachlor (mg kg⁻¹) in the marine dry fish samples collected from different markets of Dhaka District.

Heptachlor in the marine dry fish samples collected from different markets of Chattogram district are summarized in Table 7. The shrimp samples collected from Asadgonj Bazar contained heptachlor residue (0.0448 mg kg⁻¹) was above EU-MRLs [75].

Table 7 : Heptachlor residue (mg kg⁻¹) in the marine dry fish samples collected from different markets of Chattogram district.

Name of Dry fish	Area of Collection	Level of Residue (mg/kg)	MRL (mg/kg)	References
Ribbon fish	Asadgonj Bazar	0.0023	0.01	[63]
	Asadgonj Bazar	0.0012		[20]
Shrimp	Reajuddin Bazar	0.0038		[63]
	Asadgonj Bazar	0.0448		[20]
Chinese pomfret	Asadgonj Bazar	0.0053		[20]
Indian salmon	Asadgonj Bazar	0.0048		[20]
Bombay duck	Asadgonj Bazar	0.0055		[63]
	Reajuddin Bazar	0.0023		[63]
	Asadgonj Bazar	0.0012		[20]

It is observed that the range of heptachlor concentration in all marine dry fish samples were 0.0012 mg kg⁻¹ to 0.0448 mg kg⁻¹ (Fig.6). The highest concentration of heptachlor found in shrimp (0.0448 mg kg⁻¹) collected from Asadgonj Bazar and lowest in ribbon fish (0.0012 mg kg⁻¹) and bombay duck (0.0012 mg kg⁻¹) also collected from the same market.

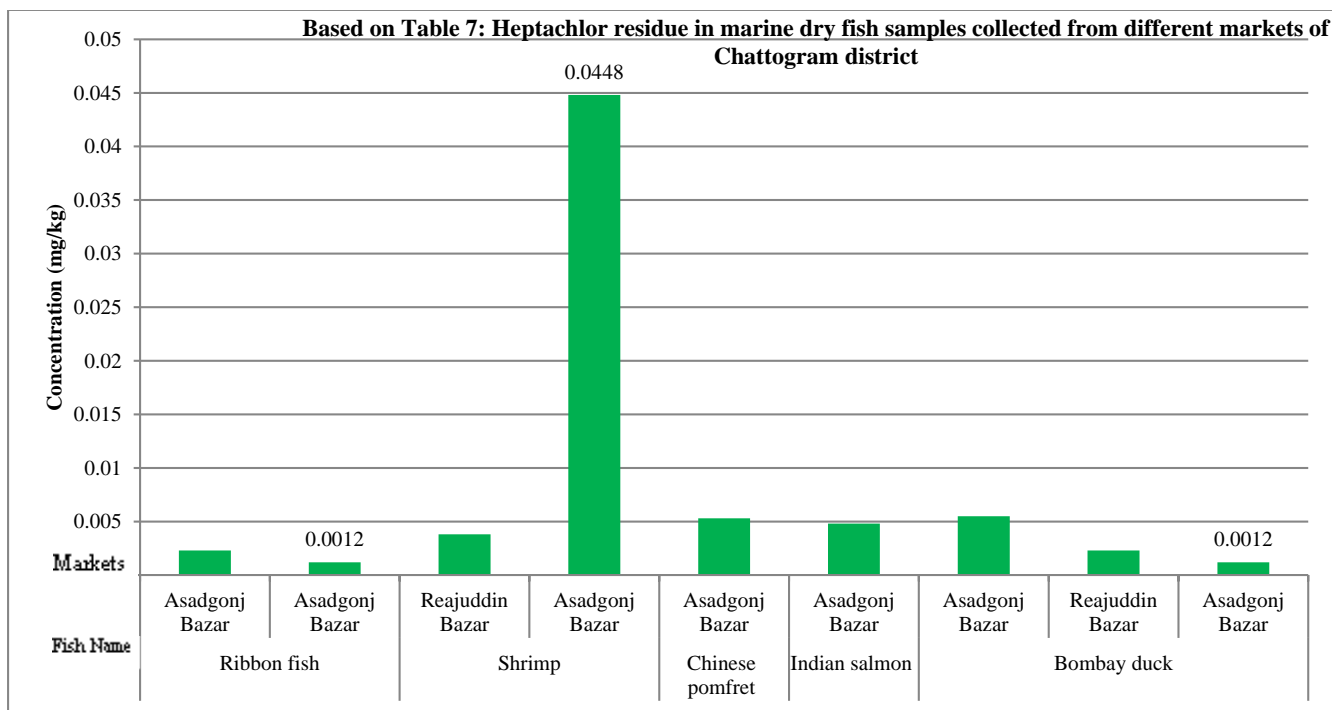


Figure 6. Level of Heptachlor (mg kg⁻¹) in the marine dry fish samples collected from different markets of Chattogram district.

Heptachlor concentration was found comparatively lowest in shrimp and highest in chinese pomfret collected from different markets of Dhaka. In Chattogram district, shrimp contained highest concentration of Heptachlor and ribbon fish and Bombay duck contained lowest concentration. Chinese pomfret and Anchovy from Dhaka and shrimp from Chattogram district contained heptachlor residue above MRL [75]. The ribbon fish samples of all places contained almost about same concentration of heptachlor. The concentration of heptachlor in dry fish compare to DDT was found too less. It is observed that all samples contained invariably banned item of DDT and heptachlor and it can assume that a composite mixture of these insecticides is used for the conservation of dry fish.

IV.C. Fresh Water Dry Fish:

Among the fresh water dry fishes, all samples collected from Dhaka district were found contaminated with DDT residue above the EU-MRLs [75] are summarized in Table 8.

Table 8: Level of pesticide residues (mg kg⁻¹) in the fresh water dry fish samples collected from different districts of Bangladesh

District Name	Name of Dry fish	Detected Pesticide	Level of Residue (mg/kg)	MRL (mg/kg)	References			
Dhaka	Chepa	DDT	0.24301	0.05	[41]			
	Puti		0.18741		[41]			
	Nona Ilish		0.09384		[41]			
	Boro Chingri		0.18348		[41]			
	Gura Chingri		0.33238		[41]			
	Kaski		0.80868		[41]			
	Chaplia		0.66572		[41]			
	Shoal		0.17723		[41]			
	Desi Chanda		0.55478		[41]			
	Ghonia		11.8805		[41]			
Bogura	Chepa	Endrin	0.08	0.01	[60]			
Dhaka	Kaski		0.105		[60]			
Bogura			0.07		[60]			
Chattogram			0.06		[60]			
Mymensingh			0.12		[60]			
Dinajpur			0.09		[60]			
Rajshahi			0.12		[60]			
Dinajpur			Chepa		0.13	[60]		
Bogura			Mola		0.12	[60]		
Chattogram			Paysha		0.07	[60]		
Bogura					0.09	[60]		
Mymensingh					0.08	[60]		
Dinajpur					0.05	[60]		
Dhaka					Chepa	Aldrin	0.284	0.01
			0.205				[59]	
	0.112	[59]						
	0.21	[59]						
	0.215	[59]						
	0.226	[59]						
	0.206	[59]						
	0.242	[59]						
	0.195	[59]						
Gazipur	Chepa	Dieldrin	0.221	[59]				
Dhaka			0.371					
Gazipur			0.527					
			0.548		[59]			

			0.778	0.01	[59]
			0.411		[59]
			0.391		[59]
Mymensingh	Shidhol	Endrin ketone	0.09	0.01	[60]
Rajshahi	Chepa	Heptachlor epoxide	0.07	0.01	[60]

The level of DDT was highest (11.8805 mg kg⁻¹) in Ghonia (*Laeo gonius*) then Kaski (*Corica soborna*), Chaplia (*Gudusia chapra*), and Desi Chanda (*Chanda ranga*) exceeded the EU-MRLs [75] are shown in Fig.7.

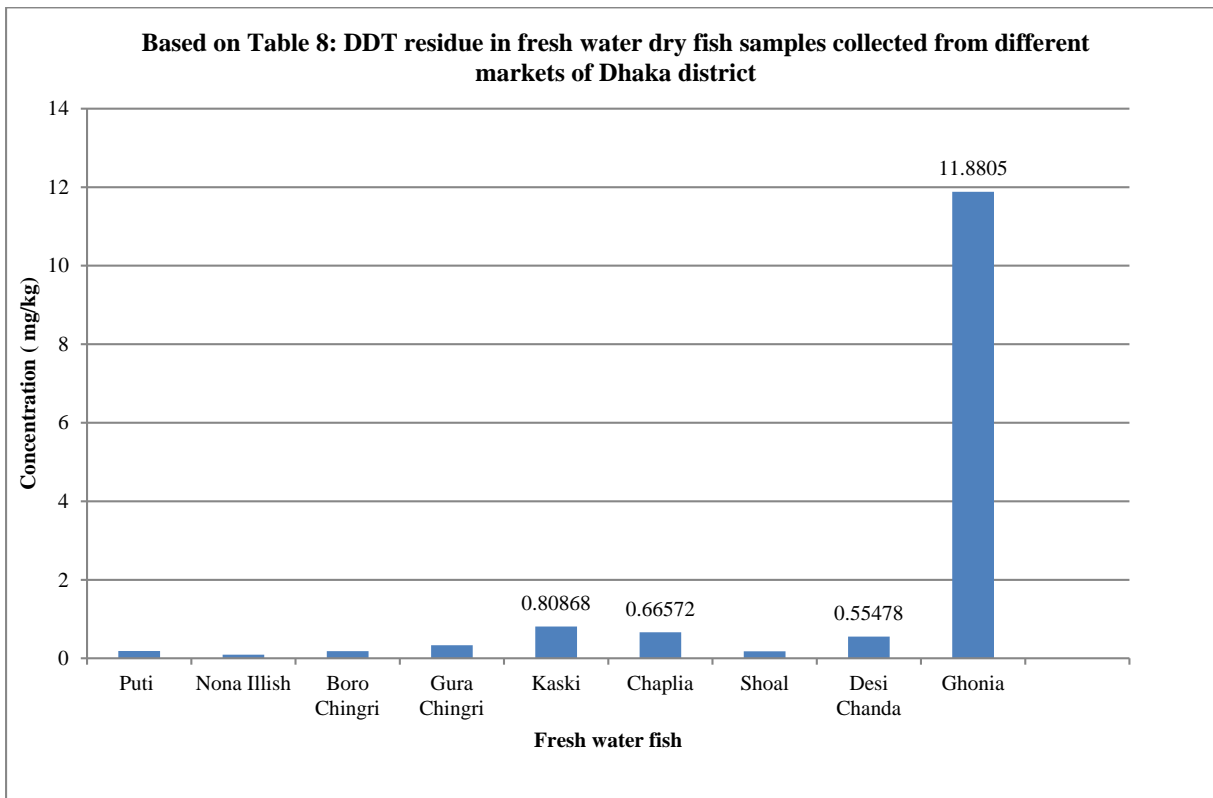


Figure 7. Level of DDT (mg kg⁻¹) in the fresh water dry fish samples collected from different markets of Dhaka district.

From Table 8, fresh water dry fishes collected from Dhaka, Gazipur, Chattogram, Mymensingh, Bogura, Rajshahi, Rangpur, Dinajpur, contained DDT, aldrin, dieldrin, endrin, endrin ketone, heptachlor, and heptachlor epoxide residue. Among them, mostly cheapa (*Puntius sophore*) collected from Dhaka and Gazipur market contaminated aldrin and dieldrin residue with above MRL. Most of the samples collected from Bogura were contaminated with endrin. The lowest concentration of endrin (0.05 mg kg⁻¹) found in paysha [60] and the highest (0.13 mg kg⁻¹) found in chepa collected from Dinajpur. Heptachlor epoxide (0.07 mg kg⁻¹) and endrin ketone (0.09 mg kg⁻¹) were detected in chepa and shidhol collected from Rajshahi and Mymensingh accordingly. Other fishes were contaminated with endrin residue at a level of above EU-MRLs.

The level of DDT in dry fish is a greater concern but more concern is such a dangerous poison is still using in our some popular dish items such as dry fish though it is banned in our country. So, health risk assessment should be done for further study. It is observed that DDT is a very slow poisoning substance [63, 76, 77]. It could be transferred from generation to generation through breast milk [78]. According to the US National Toxicological Program, it could be referred as “moderately toxic” substance or could be considered as a “moderately hazardous” substance [79]. A number of studies showed that DDT is responsible for non-allergic asthma [86] and have direct link with diabetes [80]. A study found that elevated risk of cancers of liver and biliary tract for workers who are handled DDT to control the malaria vector [81]. A number of studies argued that the accumulation of DDT in human body before puberty increases the risk of breast cancer for the women [82].

Heptachlor has been shown to bioaccumulates in fish and cattle [83]. Most of the heptachlor that is swallowed is absorbed into blood. Heptachlor can pass directly from a mother's blood to an unborn baby through the placenta [84]. Animals fed heptachlor throughout their lifetime had more liver tumors than animals that ate food without heptachlor [85]. EPA has classified heptachlor as a probable human carcinogen (B2) and established an oral cancer slope factor of 4.5 per mg/kg-day [84] and for DDT it is 0.34 per mg/kg-day.

The levels of detected OCPs found by different researchers are alarming for Bangladesh. In our study, the U.S Environmental Protection Agency’s guidelines for human health risk assessment have been taken into consideration.

IV.D. Tolerance limits and risk assessment

Risk assessment of DDT and heptachlor measured in the different marine dry fish samples collected from reviewed journals were compared with EU- MRLs (Table 9, 10)

Table 9: Estimated daily intake (EDI) (mg kg⁻¹ bwd⁻¹), hazard quotient (HQ), life time cancer risks and hazard ratios (HR) calculated for DDT concentrations detected in marine dry fish samples from Dhaka district, according to Table 3.

Cancer slope factor of DDT = 0.34 (mg/kg/day)					Non-carcinogenic Risk				Cancer Risk			
ADI= 0.02 (mg kg ⁻¹ bw d ⁻¹) MRL= 0.05 mg/kg					Hazard Quotient (HQ)		Risk (HQ≤0.2)		Hazard Ratios (HR)		Risk (HR>1)	
*Name of Dry Fish	*Area of Collection	Level of Residue (mg/kg)	EDI (Adult) (mg kg ⁻¹ bwd ⁻¹)	EDI (Child) (mg kg ⁻¹ bwd ⁻¹)	HQ(Adult)	HQ(Child)	Adult	Child	HR(Adult)	HR(Child)	Adult	Child
Rf	D N M	0.1494	6.823E-06	4.09E-05	0.00034113	0.0020468	No	No	0.00011	0.0038136	No	No
	KB	0.1316	6.01E-06	3.61E-05	0.000300487	0.0018029	No	No	9.3E-05	0.0033592	No	No
	KB	0.043158	1.971E-06	1.18E-05	9.85464E-05	0.0005913	No	No	3.1E-05	0.0011017	No	No
Sh	DNM	0.00304	1.388E-07	8.33E-07	6.94133E-06	4.165E-05	No	No	2.2E-06	7.76E-05	No	No
	KB	0.0694	3.169E-06	1.9E-05	0.000158463	0.0009508	No	No	4.9E-05	0.0017715	No	No
Bd	DNM	0.1566	7.151E-06	4.29E-05	0.00035757	0.0021454	No	No	0.00011	0.0039973	No	No
	KB	0.1823	8.325E-06	5E-05	0.000416252	0.0024975	No	No	0.00013	0.0046534	No	No
	KB	0.044395	2.027E-06	1.22E-05	0.000101369	0.0006082	No	No	3.1E-05	0.0011332	No	No
Cp	KB	0.007849	3.584E-07	2.15E-06	1.79219E-05	0.0001075	No	No	5.6E-06	0.0002004	No	No
Is	KB	0.233727	1.067E-05	6.4E-05	0.000533677	0.0032021	No	No	0.00017	0.0059661	No	No
Anc	KB	0.019416	8.867E-07	5.32E-06	4.43332E-05	0.000266	No	No	1.4E-05	0.0004956	No	No
Cr	KB	0.016404	7.491E-07	4.49E-06	3.74558E-05	0.0002247	No	No	1.2E-05	0.0004187	No	No

*DNM = Dhaka new market, KB = Karwan Bazar. Bd = Bombay duck, Cp = Chinese pomfret, Rf = Ribon fish, Sh = Shrimp, Is = Indian salmon, Anc = Anchovy, Cr = Croaker. ADI(EC 2005)[28].

Table 10: Estimated daily intake (EDI) (mg kg⁻¹ bwd⁻¹), hazard quotient (HQ), lifetime cancer risks and hazard ratios (HR) calculated for heptachlor concentrations detected in marine dry fish samples from Dhaka district, according to Table 6.

Cancer slope factor of Heptachlor = 4.5 (mg/kg/day)					Non-carcinogenic Risk				Cancer Risk			
ADI= 0.0001 (mg kg ⁻¹ bw d ⁻¹) MRL 0.01 mg/kg					Hazard Quotient (HQ)		Risk (HQ≤0.2)		Hazard Ratios (HR)		Risk (HR>1)	
*Name of Dry fish	*Area of Collection	Level of Residue (mg/kg)	EDI (Adult) (mg kg ⁻¹ bwd ⁻¹)	EDI (Child) (mg kg ⁻¹ bwd ⁻¹)	HQ (Adult)	HQ (Child)	Adult	Child	HR (Adult)	HR (Child)	Adult	Child
	D N M	0.0017	7.763E	4.66E-	0.00077633	0.004658	N	N	1.6E-05	0.0005743	N	N

Rf			-08	07	3							
	KB	0.0019	8.677E-08	5.21E-07	0.000867667	0.005206	N	N	1.8E-05	0.0006419	N	N
	KB	0.000778	3.553E-08	2.13E-07	0.000355287	0.0021317	N	N	7.3E-06	0.0002628	N	N
Sh	DNM	0.0007	3.197E-08	1.92E-07	0.000319667	0.001918	N	N	6.6E-06	0.0002365	N	N
BD	KB	0.0018	8.22E-08	4.93E-07	0.000822	0.004932	N	N	1.7E-05	0.0006081	N	N
	KB	0.001452	6.631E-08	3.98E-07	0.00066308	0.0039785	N	N	1.4E-05	0.0004905	N	N
Cp	KB	0.054753	2.5E-06	1.5E-05	0.02500387	0.1500232	N	N	0.00051	0.0184979	N	N
IS	KB	0.002532	1.156E-07	6.94E-07	0.00115628	0.0069377	N	N	2.4E-05	0.0008554	N	N
Anc	KB	0.018306	8.36E-07	5.02E-06	0.00835974	0.0501584	N	N	0.00017	0.0061845	N	N
Cr	KB	0.014856	6.784E-07	4.07E-06	0.00678424	0.0407054	N	N	0.00014	0.005019	N	N

*DNM = Dhaka new market, KB = Karwan Bazar. Bd = Bombay duck, Cp = Chinese pomfret, Rf = Ribon fish, Sh = Shrimp, Is = Indian salmon, Anc = Anchovy, Cr = Croaker. ADI (EC 2005)[28].

Some of the analyzed marine dry fish samples collected from Dhaka were contaminated with DDT and heptachlor with mean concentrations exceeding the EC guideline limits (Table 9, 10). The EDIs and health risk estimates for both cancer and non-cancer risks were calculated for DDT and heptachlor for dry marine fish samples which were also collected from Chattogram and Khulna district of Bangladesh and found to indicate negligible adverse health effects $HI < 1 (\sum HQ = HI)$ and cancer risk ($HR < 1$) both for adult and child like Dhaka district.

Table 11. Estimated daily intake (EDI) ($\text{mg kg}^{-1} \text{ bwd}^{-1}$), hazard quotient (HQ), lifetime cancer risks and hazard ratios (HR) calculated for endrin, aldrin, dieldrin, endrin ketone, heptachlor epoxide concentrations detected in fresh water dry fish samples from different districts of Bangladesh according to Table 8.

*District name	Name of Dry fish	Name of pesticide	Level of Residue (mg/kg)	ADI (mg /kg bw /d)	Cancer slope factor (mg/kg/day)	EDI (Adult) (mg /kg bw /d)	EDI (Child) (mg /kg bw /d)	Non-carcinogenic Risk				Cancer Risk					
								Hazard Quotient (HQ)		Risk (HQ≤0.2)		Hazard Ratios (HR)		Risk (HR>1)			
								HQ (Adult)	HQ (Child)	Adult	Child	HR (Adult)	HR (Child)	Adult	Child		
Dhk	Chepa	DDT	0.243	0.02	0.34	1E-05	7E-05	0.0006	0.0033	No	No	2E-04	N0	No	No		
Dhk	Puti		0.1874			8.5583E-06	5.13503E-05	0.00042792	0.002567517	No	No	0.00664416	0.23919	No	No	No	No
Dhk	Nona Illish		0.0938			4.2853E-06	2.57122E-05	0.000214268	0.001285608	No	No	0.00332687	0.119767	No	No	No	No
Dhk	Boro Chingri		0.1834			8.3789E-06	5.02735E-05	0.000418946	0.002513676	No	No	0.00650483	0.234174	No	No	No	No
Dhk	Gura Chingri		0.3323			1.5178E-05	9.10721E-05	0.000758934	0.004553606	No	No	0.01178372	0.424214	No	No	No	No
Dhk	Kaski		0.8086			3.6929E-05	0.000221578	0.001846486	0.011078916	No	No	0.02866977	1.032112	No	Yes	No	Yes
Dhk	Chaplia		0.6657			3.0401E-05	0.000182407	0.001520061	0.009120364	No	No	0.02360148	0.849653	No	No	No	No
Dhk	Shoal		0.1772			8.0935E-06	4.8561E-05	0.000404675	0.002428051	No	No	0.00628326	0.226197	No	No	No	No
Dhk	Desi Chanda		0.5547			2.5335E-05	0.00015201	0.001266748	0.007600486	No	No	0.01966837	0.708061	No	No	No	No

Dhk	Ghonia		11.8805			0.000542543	0.003255257	0.027127142	0.16276285	Yes	Yes	0.42119409	15.16299	No	Yes
Bog	Chepa	Endrin	0.08	0.0002	N/A	4E-06	2E-05	0.0183	0.1096	No	No	N/A			
Dhk	Kaski		0.105			5E-06	3E-05	0.024	0.1439	No	No				
Bog	Kaski		0.07			3E-06	2E-05	0.016	0.0959	No	No				
Ctg	Kaski		0.06			3E-06	2E-05	0.0137	0.0822	No	No				
Msh	Kaski		0.12			5E-06	3E-05	0.0274	0.1644	No	No				
Din	Kaski		0.09			4E-06	2E-05	0.0206	0.1233	No	No				
Raj	Kaski		0.12			5E-06	3E-05	0.0274	0.1644	No	No				
Din	Chepa		0.13			6E-06	4E-05	0.0297	0.1781	No	No				
Bog	Mola		0.12			5E-06	3E-05	0.0274	0.1644	No	No				
Ctg	Mola		0.07			3E-06	2E-05	0.016	0.0959	No	No				
Bog	Paysha		0.09			4E-06	2E-05	0.0206	0.1233	No	No				
Msh	Paysha		0.08			4E-06	2E-05	0.0183	0.1096	No	No				
Din	Paysha		0.05			2E-06	1E-05	0.0114	0.0685	No	No				
Dhk	Chepa	Aldrin	0.284	0.0001	17	1E-05	8E-05	0.1297	0.7782	No	Yes				
Dhk			0.205			9E-06	6E-05	0.0936	0.5617	No	Yes	0.007	0.3	No	No
Dhk			0.112			5E-06	3E-05	0.0511	0.3069	No	Yes	0.004	0.1	No	No
Dhk			0.21			1E-05	6E-05	0.0959	0.5754	No	Yes	0.007	0.3	No	No
Dhk			0.215			1E-05	6E-05	0.0982	0.5891	No	Yes	0.008	0.3	No	No
Gaz			0.226			1E-05	6E-05	0.1032	0.6192	No	Yes	0.008	0.3	No	No
Gaz			0.206			9E-06	6E-05	0.0941	0.5644	No	Yes	0.007	0.3	No	No
Gaz			0.242			1E-05	7E-05	0.1105	0.6631	No	Yes	0.009	0.3	No	No
Gaz			0.195			9E-06	5E-05	0.0891	0.5343	No	Yes	0.007	0.2	No	No
Gaz			0.221			1E-05	6E-05	0.1009	0.6055	No	Yes	0.008	0.3	No	No
Dhk	Chepa	Dieldrin	0.371	0.0001	16	2E-05	0.0001	0.1694	1.0165	No	Yes	0.012	0.5	No	No
Gaz			0.527			2E-05	0.0001	0.2407	1.444	No	Yes	0.018	0.7	No	No
Gaz			0.548			3E-05	0.0002	0.2503	1.5015	Yes	Yes	0.018	0.7	No	No
Gaz			0.778			4E-05	0.0002	0.3553	2.1317	Yes	Yes	0.026	1	No	No
Gaz			0.411			2E-05	0.0001	0.1877	1.1261	No	Yes	0.014	0.5	No	No
Gaz			0.391			2E-05	0.0001	0.1786	1.0713	No	Yes	0.013	0.5	No	No
Msh	Shidhol	Endrin ketone	0.09	0.0002	N/A	4E-06	2E-05	0.0206	0.1233	No	No	N/A			
Raj	Chepa	Heptachlor epoxide	0.07	0.0001	9	3E-06	2E-05	0.032	0.1918	Yes	Yes	0.001	0	No	No

*Dhk = Dhaka, Bog = Bogura, Ctg = Chattogram, Msh = Mymensingh, Din = Dinajpur, Raj = Rajshahi, Gaz = Gazipur.

Data derived from collected fresh water dry fish samples are used for risk assessment presented in Table 11. The estimated EDIs for most analytes were below their respective ADI values set by the US EPA for adult but for child notable exceptions included DDT, aldrin, dieldrin and endrin which had higher EDIs than their respective ADIs. Non-cancer risk estimates showed that aldrin, dieldrin and endrin were present in dry fish chepa that could pose immediate dietary risk for adult and acute risk for child, notable exception for DDT in ghonia fish estimated immediate dietary risk for both adult and child. This is particularly so for which had $HQ > 0.2$ at concentrations ranging between 0.548 - 0.0778 mg kg⁻¹ for dieldrin, which is exceeded the acceptable limit $HI < 1$ ($\sum HQ = HI$) for

Adult. So, adult are vulnerable to health risk due to dieldrin consumption through dried fish chepa. For child, this range is far more wider for aldrin (0.112- 0.242 mg kg⁻¹) and for dieldrin (0.371 - 0.778 mg kg⁻¹). For child, both aldrin and dieldrin pesticide consumption through dry fish cheap exceeded the threshold limit of 1(HI<1) indicating adverse potential dietary risk. Now a further detailed risk assessment or risk management measures to be undertaken.

Cancer risk estimates (hazard ratios and lifetime cancer risks) for aldrin, dieldrin, heptachlor epoxide are presented in Table 11. Hazard ratios (HR) for aldrin, dieldrin, heptachlor epoxide not exceeded threshold values. So for child, no potential cancer risk from the consumption of locally sourced fresh water dried fish chepa and shidhol. DDT in ghonia fish shows potential cancer risk for child. Child is more vulnerable cancer risk then the adult.

V. Conclusion

Present study revealed that dry fish samples collected and analyzed by several researchers from Dhaka, Gazipur, Chattogram, Khulna, Mymensingh, Bogura, Rajshahi, Rangpur and Dinajpur were found to be contaminated with different concentration of DDT, heptachlor, aldrin, dieldrin, endrin, endrin ketone, and heptachlor epoxide indicating unrestricted and unplanned use of them without considering their health hazard. The detection of multiple pesticides in dried fish of retail markets revealed that fishers might use more than one pesticide in dried fish. Another explanation could be that one pesticide might be used by producers and another might be used by dried fish value chain vendors for long-term preservation of fish. Continual development and modification of pesticide legislation and policies should be done based on modern and renewable pesticide databases. The government of Bangladesh should take all the necessary steps to combat the situation; it can be the steps to implement the legislation and improving the awareness of the related people through some program, public education campaigns and announcement of harmful role of these insecticides.

VI. Acknowledgements

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