Organochlorine concentrations in aquatic organisms from different trophic levels of the Sundarbans mangrove ecosystem and their implications for human consumption

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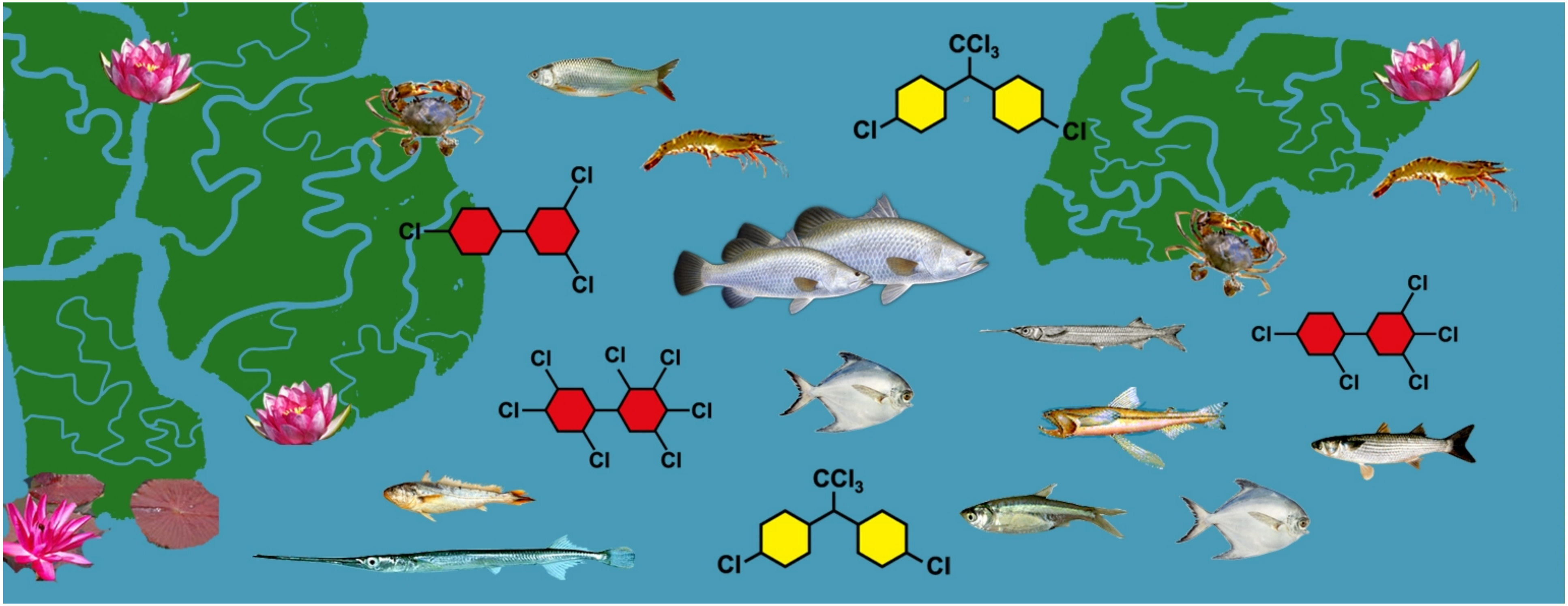
Highlights

- DDTs and PCBs were identified in organisms from the Sundarbans mangrove ecosystem
- Levels found were lower than those in wildlife from other mangrove ecosystems
- Levels in edible fish are not considered to pose a risk for human consumption
- Food chain length in the mangrove ecosystem was found to be remarkably short
- No relationship was found between organochlorine concentrations and trophic levels



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Abstract

The Sundarbans, a highly biodiverse tropical ecosystem stretching across India and Bangladesh, is also the largest mangrove forest in the world. Organochlorine compounds (OCs) have been extensively used for agriculture and sanitary purposes in the region. OCs can accumulate in biological tissues and biomagnify in organisms through food webs, for which reason they reach high concentrations in top predators. Because marine food webs are long and marine predators are extensively used in the region as human food, assessment of potential health-related risks caused by OC pollution is in order. This study is the first to determine the concentration of PCBs in fish and crustaceans from the Sundarbans mangroves, their accumulation trends through the food web, and the potential toxicological risk that their consumption poses to humans. DDT concentrations, which had already been assessed in the region, were also determined. The median concentrations ranged from below detection limits to 176.3 ng g^{-1} lipid weight for tDDT and 30,982 ng g^{-1} for PCBs. Overall, these concentrations were lower than those usually observed in other regions of the world, apparently as a result of the interplay of several factors: low environmental organochlorine inputs, the physical and climatic characteristics of an ecosystem dominated by high temperatures in a highly flushed ecosystem that dilutes and rapidly disperses pollutants, and the comparatively short food chain lengths that, similarly to other mangrove ecosystems, characterize the Sundarbans. Organochlorine concentrations were 2-3 orders of magnitude lower than commonly accepted tolerance levels, so their consumption do not pose a sensible risk to the population. However, concentrations of DDT in dry fish from retail markets were higher because this compound is used for pest control during fish processing. Potential risks involved in this practice likely outweigh potential benefits, so it is recommended that this compound is substituted by less hazardous alternatives.

Capsule: Organochlorine concentrations in aquatic organisms from the Sundarbans mangrove ecosystem are low, not related to trophic level, and do not pose a risk to human consumption

Keywords: Bangladesh; POP, trophic web; human consumption; food safety; mangrove ecosystem

1. Introduction

The Sundarbans is a highly productive ecosystem spreading across India and Bangladesh. It is the largest continuous mangrove forest in the world and is extremely rich in biodiversity (Gopal and Chauhan, 2006). For these reasons it has been declared a World Heritage Site by UNESCO in 1987. This mangrove forest lies on the extensive delta formed by the confluence of three major river systems - the Ganges, Brahmaputra, and Meghna- at the northern apex of the Bay of Bengal (Fig. 1). It comprises coastal mangroves and marsh islands where numerous marine species spawn and breed (Nagelkerken et al., 2008). All the rivers and organisms in this region are subjected to the ebb and flow of tidal flooding, which constantly renews the habitat. For centuries, the delta has been exploited for agricultural (Getzner and Islam, 2013) and fisheries purposes (Mustafa, 2009). However, in recent decades overfishing and overexploitation of plant and wildlife species are placing great stress on the viability of the ecosystem (Islam and Haque, 2004).

Recent studies indicate that the Sundarbans aquatic ecosystem is reeling from the effects of indiscriminate anthropogenic activities that result in poor water quality and the accumulation of chemical contaminants, such as heavy metals (Mitra et al, 2011, 2012; Borrell et al, 2016), organochlorine pesticides, polychlorinated biphenyls (PCBs) (Ahmad et al., 1996; Sarkar et al., 2008a; Binelli et al., 2009; Yadav et al., 2015) and polycyclic aromatic hydrocarbons (Guzzella et al., 2005).

Organochlorine compounds (OCs) are synthetic contaminants known to be persistent and to adversely impact ecosystem processes and biodiversity (Islam and Tanaka, 2004; Jepson et al., 2016). In the Indian subcontinent, OCs have been extensively used both as industrial compounds (e.g., PCBs) and as pesticides in agricultural applications and against the malaria vector (e.g., DDT or dichlorodiphenyltrichloroethane) due to their effective results and low economic cost (Pandit et al., 2001; Sarkar et al., 2008a). The locally active shipbreaking industry is also a main generator of PCB pollution, with up to 0.25-0.8 metric tons of PCBs released per scrapped ship (Cheng et al., 2015). Through discharge and surface runoff from sources, as well as wet and dry atmospheric deposition, the released OCs enter surrounding water bodies, where they adhere to organic particles in suspension or associate with sediments (Sarkar, 2008b; Binelli, 2009; Ahmed et al., 2015). Ultimately, OCs disperse in the environment and can cause global contamination of wildlife populations (Sarkar et al., 2008a), including edible fish (Jabber et al., 2001;

Hasan et al., 2014) and even reaching humans, as shown by their presence in human breast milk (Someya et al, 2010).

In the Sundarbans, PCBs and DDTs are known to be widely present in sediments (Bhattacharya et al., 2003; Guzzella et al., 2005), so the progressive industrialization and increased use of marine organisms as human food demands rigorous control over their concentration throughout the ecosystem. The properties of OCs facilitate their accumulation in lipid-rich tissues and their biomagnification through the food web (e.g., Borgå et al., 2001; Hoekstra et al., 2003), causing adverse biochemical and physiological effects to top predators (e.g., Borrell et al., 1996; Troisi et al., 2001), as well as the humans that consume these organisms.

Information about the occurrence of OC pollutants in aquatic organisms in the eastern region of the Sundarbans mangrove (Bangladesh) appears to be non-existing. Particularly, there appears to be a complete absence of data on PCBs in tissues from edible fish and crustaceans. To fill this gap, this study aims to: 1) determine the concentrations of DDT and PCB in representative marine organisms of this ecosystem and assess the potential for biomagnification of these compounds through parallel determination of nitrogen stable isotope ratios of the analysed organisms, and 2) evaluate the toxicological risk to humans resulting from the consumption of aquatic species from this region.

2. Materials and methods

2.1 Sample collection

In December 2011, 14 different species, 10 fish, 2 crustaceans and 2 plants (Table 1), were collected from the Sundarbans mangrove of Bangladesh (Fig. 1). The selection of species was based on their relevance to the mangrove ecosystem and in their use as food for humans. Moreover, species were selected to create a representation of the food web that enabled the assessment of biomagnification patterns.

The fish and crustaceans (5 individuals per species) were obtained from the local market at Khulna (site 1, Fig. 1), a town located at the border of the Sundarbans, after ensuring that their origin was the Sundarbans mangrove. All specimens were identified to the level of species and their total body length measured. This information and the inferred habitats and typical prey of each species are detailed in Table 1.

Table 1. Total length and standard deviation (SD) of the sampled specimens, and literature-derived biological lengths, feeding habitats and food items (in other regions) of the sampled species extracted from FishBase (2019) (fish), from FAO Fisheries and Aquaculture Department (2018) (*Penaeus monodon*) and from Davie and Mann (1988) (*Scylla serrata*). * In *Scylla serrata* carpace width, instead of length, is shown.

				From the literature				
Species	**Common name**	**n**	**Length (mean±SD (cm)**	**Maturity length (cm)**	**Common length (cm)**	**Maximum length (cm)**	**Habitat**	**Food items**
Scylla serrata	Mangrove crab	5	11.2±0.8*	12*	14*	19*	Mangroves in estuaries and sheltered coastal habitats. in soft muddy bottoms	Zoobentos; molluscs and small crabs
Penaeus monodon	Giant tiger prawn	5	18±0		16	33	Bottom mud, sand (depth range 0 - 110 m). Estuarine (juveniles) and marine (adults)	Molluscs, small crustaceans
Mugil cephalus	Flathead mullet	5	18±0.8	35.4	50	100	Coastal waters and estuaries (usually in schools over sand or mud bottom)	Detritus, micro-algae and benthic organisms
Amblypharyngodon mola	Mola carplet	5	4±0.6	6		20	Rivers, canals, ponds and inundated fields	Detritus, zooplancton
Harpadon nehereus	Bombay duck	5	20±1	13-	25	40	Deep water offshore on sandy mud bottoms shallower than 50 m depth and deltas of rivers	Nekton, finfish, bony fish
Tenualosa ilisha	Ilish	5	24.2±0.8	41.5	36	60	Schooling in coastal waters and ascending rivers for 50-100 km	Plankton
Lates calcarifer	Barramundi	5	85±2.5	45	150	200	Coastal waters, estuaries and lagoons (depth range 10 - 40 m)	Fish and crustaceans
Panna microdon	Picnic seabream	5	24.4±1.8		20	30	Shallow coastal waters and estuaries; young and juveniles occur in mangrove swamps	Not known
Strongylura leiura	Panna croaker	5	22.6±1.3		35	100	Coastal waters and estuaries. Larvae and early juveniles in mangroves (depth range 0 - 3 m)	Small fish and crustaceans
Acanthopagrus berda	Banded needlefish	5	26.5±0.9	21	35	90	Marine; freshwater; brackish; demersal (depth range? - 50 m)	Invertebrates and small fish
Hyporhamphus limbatus	Congaturi halfbeak	5	13.5±0.4	9	13	35	Coastal waters and at surface levels of tidal freshwaters and brackish estuaries	Mainly insects
Pampus argenteus	White pomfret	5	21.7±1.6	25.3	30	60	Inshore, usually in schools over muddy bottoms (depth range 5 - 110 m)	Ctenophores, salps, medusae, and other zooplankton groups

Muscle samples were taken from all individuals except for *Amblypharyngodon mola* which, due to its small size (< 4 cm), could not be properly dissected and therefore its whole body was used for the analyses. The samples of plants were collected directly in the field by us (site 2, Fig. 1). All samples were oven-dried (40°C, 72 hours) *in situ* in a portable food dehydrator (Excalibur Food Dehydrator). Once in the laboratory, the samples were stored at -20°C until analysis.

2.2 Stable isotope ratios of Nitrogen

Stable isotopes of Nitrogen were determined to assign a trophic level (TL) to each species (see below). To do so, approximately 1 g of the dried sample was homogenized and lipid extracted using sequential soakings in a chloroform:methanol (2:1) solution (Murphy, 1972). After these treatments and subsequent oven-drying, dilapidated subsamples of approximately 0.5 mg were placed into tin buckets, which were crimped for combustion. Isotope analyses were performed by means of elemental analysis-isotope ratio mass spectrometry using a Thermo Finnigan Flash 1112 (CE Elantech, Lakewood, NJ, USA) elemental analyser, coupled to a Delta C isotope ratio mass spectrometer via a CONFLO III interface (Thermo Finnigan MAT, Bremen, Germany).

Stable isotope abundances were expressed in delta (δ) notation, where the relative variations of stable isotope ratios are calculated in per mil (‰) deviations from predefined international standards according to the equation:

$$\delta^{15}N = [(^{15}N/^{14}N \text{ sample}/^{15}N/^{14}N \text{ standard}) - 1] \times 1000$$

The standard reference material was nitrogen gas in the atmosphere.

The isotopic ratio mass spectrometry facility at the laboratory of the Centres Científics i Tecnològics of the University of Barcelona (Spain) applies international isotope secondary standards of known R ratios supplied by the International Atomic Energy Agency (IAEA, Vienna). Secondary standards for nitrogen of known $^{15}N/^{14}N$ ratios were $(NH_4)_2SO_4$ (IAEA-N-1, $\delta^{15}N$ = +0.4‰ and IAEA-N-2, $\delta^{15}N$ = +20.3‰), and KNO_3 (IAEA-NO-3, $\delta^{15}N$ = +4.7‰). All of the standards were inserted in the analytical runs every 12 samples to calibrate the system and compensate for any drift over time. Replicate assays of standard materials indicated $\delta^{15}N$ measurement errors of ±0.3‰.

2.3 Trophic level calculation for biomagnification assessment

Based on the process of ^{15}N enrichment in consumers over their prey (Cabana and Rasmussen, 1996, Post, 2002), the trophic position of each of the sampled organisms was determined according to their relative abundance of ^{15}N to ^{14}N ($\delta^{15}N$). We used the plants as the baseline for TL $\delta^{15}N$ estimations (mean value of *Ceriops decandra* and *Nymphaea pubescens;* $\delta^{15}N_{baseline}$ = 3.69‰) (TL = 1). As the mean enrichment of $\delta^{15}N$ per trophic level is 3.4 (Post, 2002), trophic levels (TLs) for each species were estimated from raw $\delta^{15}N$ values using the following equation:

$$TL\ \delta^{15}N = TL_{baseline} + (\delta^{15}N_{species} - \delta^{15}N_{baseline})/3.4$$

2.4 Organochlorine compounds

To analyse fish and crustaceans for OCs, approximately 1 g of the dried tissue sample was ground with anhydrous sodium sulphate using a mortar. The mixture was extracted with n-hexane for 4 hours in a Soxhlet apparatus with 125 ml of capacity. The solution obtained was concentrated to 40 ml. A portion of this extract (10 ml) was used to gravimetrically determine the quantity of extractable fat per gram of the dried sample. The rest of the solution was mixed with sulphuric acid for the clean-up, following the procedures described by Murphy (1972), and the resulting extract was concentrated to 1 ml, centrifuged for five minutes and prepared to be injected in the Gas chromatography–mass spectrometry.

GC–MS/MS spectra were obtained on a Thermo Trace GC Ultra system (Thermo Scientific, Waltham, MA, USA) equipped with a TRB5-MS column (30 m × 0.25 mm i.d. × 0.25 μm film thickness) operating with helium as the carrier gas, coupled to a Thermo ITQ 900 mass spectrometer (MS). The GC injector was operated in a pulsed splitless mode. The volume of each injection was 1 μl. The injector temperature was 280 °C and the GC oven was programmed to hold 90 °C for 1 min, then raise the temperature at 6 °C/min to 300 °C, which was held for 5 min. The MS was operated with the ion source at 200 °C, scanning from *m/z* 50 to 550.

The samples were analysed for the following compounds: p,p'-DDE (dichlorodiphenyldichloroethylene), p,p'-DDD (dichlorodiphenyldichloroethane), o,p'-DDT and p,p'-DDT (dichlorodiphenyltrichloroethanes) and polychlorinated biphenyls (PCBs). The tDDT concentration was calculated as the sum of the four DDT compounds. The total PCB concentration (PCB) was calculated as the sum of the 13 congeners known as IUPAC# 128, 138, 149, 153, 170, 174, 177, 180, 183, 187, 194, 196, and 201. Identification and quantification of the individual compounds were performed by comparison with external reference standards calibrated with a six-point calibration curve encompassing the entire concentration range. The linear calibration curves were constructed by analyzing standard solutions of different concentrations including 1, 10, 50, 100, 500 ng ml^{-1} for PCB congeners: 138, 153, 170 and 180, and 1, 3, 5, 10, 20, 50 ng ml^{-1} for DDTs and the rest of PCB congeners. Analyses were run in a series of five samples followed by one blank with no matrix to evaluate background contamination. The recoveries of the OCs were calculated by adding 50 ng g^{-1} (for PCBs :138, 153, 170, and 180) and 10 ng g^{-1} (for DDTs and the rest of PCBs) to 12 homogenised dry muscle replicates of the species *Panna microdon*. Recovery levels ranged from 82% to 101% with a relative standard deviation below 20%. OCs sample concentrations were not corrected for recovery as calibration standards followed the same extraction procedure as samples. Limit of detection (LOD) for each compound was calculated based on a signal to noise ratio of 3:1 from the spiked sample. LOD varied with the compounds, but typically ranged from 0.005 to 0.05 ng g^{-1} lipid weight basis (l.w.).

Since OCs are highly apolar, concentrations in this paper are expressed in ng g^{-1} l.w. For comparative purposes the l.w. concentrations can be converted to dry weight basis (d.w.) through the lipid content. To transform from d.w. to wet weight (w.w.) throughout the manuscript and table 4, a 74% moisture value was used for all fish species (Huss, 1995).

3. Results

3.1. Isotopic compositions of organisms and trophic level

Fig. 2 shows for each species the $\delta^{15}N$ values and the trophic levels (TL $\delta^{15}N$) calculated according to the formula used by Post (2002) (Borrell et al., 2016), as well as the trophic levels from other sources to allow proper comparison. $\delta^{15}N$ values ranged from 3.6‰ in the primary producer *Ceriops decandra* to 11‰ in *Pampus argenteus.* The distributions of $\delta^{15}N$ values indicated that the organisms analysed consisted of three trophic levels. The two plants (*Ceriops decandra* and *Nymphaea pubescens)* were in the first trophic level. The two species of crustaceans (*Scylla serrata* and *Penaeus monodon*) and two species of fish *(Mugil cephalus* and *Amblypharyngodon mola)* were at the second level of production (secondary producers). Six species of fish (*Harpadon nehereus, Tenualosa* *ilisha, Lates calcarifer, Acanthopagrus berda, Panna microdon* and *Strongylura leiura)* were between the second and third level of production, and two species of fish (*Hyporhamphus limbatus* and *Pampus argenteus*) were at the third level of production in the food web.

3.2. Organochlorine concentrations in organisms

The concentrations of OCs found in different species are shown in Table 2. In several cases, concentrations were very low and below analytical detection limits (<d.l.). Because some specimens presented extreme OC concentrations the results are displayed as median values, which are not so skewed as means by extremely large or extremely small values. The median concentration ranges were <d.l.-176 ng g^{-1} l.w. for tDDT and <d.l. -276 ng g^{-1} l.w. for PCB. The within species variability was high, and several species contained outliers for high concentrations of tDDT and PCB (Fig. 3). For example, PCB levels in *H. nehereus* were below detection limits in all but two specimens, which showed concentrations of 15,376 and 227 ng g^{-1} l.w.

Table 2. Median, maximum and minimum concentrations of tDDT and PCB in different species from the Bangladesh Sundarbans expressed on a lipid weight basis (<d.l.: below detection limits).

	% lipids relative to d.w.		**tDDT (ng g^{-1} l.w.)**			**PCB (ng g^{-1} l.w.)**		
	Mean	SD	Median	Max.	Min.	Median	Max.	Min.
Scylla serrata	1.76	0.24	176.3	1,559	<d.l.	275.9	10,710	<d.l.
Penaeus monodon	2.33	0.10	<d.l.	<d.l.	<d.l.	<d.l.	6,637	<d.l.
Mugil cephalus	9.36	5.03	80.8	519.0	<d.l.	206.1	16,062	<d.l.
Amblypharyngodon mola	25.14	1.19	<d.l.	<d.l.	<d.l.	<d.l.	<d.l.	<d.l.
Harpadon nehereus	12.97	4.48	<d.l.	352.1	<d.l.	<d.l.	15,376	<d.l.
Tenualosa ilisha	25.62	9.21	28.6	57.3	10.7	17.2	763.7	<d.l.
Lates calcarifer	3.06	2.13	<d.l.	112.7	<d.l.	74.7	2,649	<d.l.
Acanthopagrus berda	9.73	3.61	<d.l.	<d.l.	<d.l.	<d.l.	<d.l.	<d.l.
Panna microdon	3.95	2.49	<d.l.	<d.l.	<d.l.	<d.l.	<d.l.	<d.l.
Strongylura leiura	5.62	0.72	<d.l.	232.6	<d.l.	<d.l.	398.6	<d.l.
Hyporhamphus limbatus	9.10	1.51	60.0	351.9	<d.l.	<d.l.	1,150	<d.l.
Pampus argenteus	6.41	4.54	<d.l.	819.8	<d.l.	127.8	30,982	<d.l.

The box plots distribution graphs for PCB and tDDT concentrations are shown in Fig. 3, where the species are ranked according to their trophic level.

No relationship was observed to occur between OCs concentrations and trophic level. OCs were not detected in three species situated at different trophic levels (*A. mola, P. microdon and S. leiura)*, and the species that showed the highest pollutant levels were indeed situated in the lowest position in the trophic web as estimated from $\delta^{15}N$ values. Thus, expressed in lipid weight basis and without considering the outliers, *S. serrata* showed the highest median tDDT and PCB concentrations (Table 3; Fig. 3).

It is noteworthy that he only DDT compound found in all organisms except in *T. Ilisha* was p,p'-DDE. *T. Ilisha* showed to also have p,p'-DDT and o,p'-DDE in minor concentrations. Regarding PCBs, congener 180 was the most abundant followed by congeners 170, 187, 153 and 138. This is according expectations because these highly chlorinated congeners combine a relative high abundance in commercial formulations with resistance to degradation and, as a consequence, they are often the most abundant in natural ecosystems (*e.g.* McFarland and Clarke 1989; Batang et al., 2016). Similarly, to tDDTs, no relationship between congener concentration and trophic level was observed.

4. Discussion

4.1. Organochlorine concentrations in organisms

Among environmental pollutants, PCBs and DDTs have received focused attention because of their toxicity and widespread occurrence in high concentrations in the environment even in remote areas (Bonito et al., 2016; Reijnders et al., 2018). The manufacture of these compounds peaked between 1960 and the late 1970s and, although they are still limitedly used in certain areas, their overall production and usage was banned in most countries in the late 1970s (Iwata et al., 1994; Aguilar and Borrell, 2005). However, their chemical stability and the slow biodegradation of many of their forms have transformed these compounds into ubiquitous xenobiotic pollutants. This is particularly true for marine environments, in which these pollutants have been found from the Arctic to the Antarctic and from the intertidal to the abyssal regions (Islam and Tanaka, 2004).

In Bangladesh, DDTs were officially used in agriculture from the mid-1950s until 1991 but were banned in late 1993 (Matin et al., 1998). However, its illegal use by farmers continued and, indeed, increased since the year 2000 (Bergkvist et al. 2012). Similar situation occurs in neighbouring India, which is currently the largest producer of DDT and allows its use for disease vector control (Van den Berg, 2009) although a fraction of the production also goes into agriculture (Kaushik et al, 2010, Yadav et al, 2015) despite the worldwide prohibition of the use of this compound as pesticide in 1996 (Battu et al., 2004). The commercial formulations of DDT are largely composed of p,p'DDT, while DDE (mostly as p.p'DDE) only represents a minimal fraction of the pesticide. The occurrence of DDE in the tissues of living organisms originates from environmental or physiological degradation of the parent DDT forms. As a consequence, when interpreting the pollutant load of an organism it is generally accepted that a large contribution of DDE to the total tissue burden of DDT reflects an old exposure to the pesticide, while a low contribution reflects exposure to DDT recently entering the environment (Aguilar, 1984; Borrell and Aguilar 1987). In the fish samples examined in this study, p,p'DDE was almost the only form of DDT found, indicating negligible use of this pesticide in recent times.

Usage of PCBs is also banned in Bangladesh but these compounds are still used in the electric energy sector under different trade names (Bergkvist et al., 2012) and it is known that substantial amounts of these compounds are released into the environment as a result of shipbreaking, an industry that remains very active in the region (Cheng et al., 2015). However, similarly to DDTs, the congeners that contribute largely to the PCBs mix are those that combine large presence in commercial formulations with high environmental persistence. This again indicates that inputs into the environment of "fresh" PCBs are negligible.

Bonito et al. (2016) reviewed the levels of persistent pollutants in marine fish worldwide and found that fish from the Indian Ocean had lower mean concentrations of both tDDT (3.7 ng g^{-1} w.w) and PCB (9.3 ng g^{-1} w.w.) than fish from the other four global regions studied (*i.e.* East Pacific Ocean, West Pacific Ocean, Atlantic Ocean and Mediterranean Sea). The present study was conducted in the Sundarbans mangrove ecosystem, where the river system runs through its final stage. Previous organochlorine concentration data on aquatic organisms in the region were available for DDTs, but not for PCBs. The results here obtained are consistent with those found in sediments along the lower stretch of the Hugli (Guzzella et al., 2005), a river that distributes the waters of the Ganges and one of the great rivers that flows into the Sundarbans, as well as with those from the sediments around Sagar Island, in the southern part of the mangrove estuary (Bhattacharya et al., 2003). All these studies are coincidental in showing that the PCB and tDDT levels found in fish and crustaceans from the Sundarbans are generally lower than those usually observed in other regions of the world, particularly in temperate latitudes (Bonito et al., 2016). Thus (Table 4), tDDT levels are lower than those found along the Ganges river (Sinha and Loganathan, 2015) and in marine locations in the Bay of Bengal (Shailaja and Singbal, 1994; Jabber et al., 2001; Das et al., 2002; Das and Das, 2004), but similar to those found in marine fish off Madras and other southern regions of the Indian subcontinent (Rajendran et al., 1992). It is noteworthy that the tDDT concentrations found in fish from the South Patches marine fishing ground in the Bay of Bengal (see Fig. 1 for the location) are between two and three orders of magnitude higher than those observed in the Sundarbans (Table 4) despite this being an offshore location. While the reason for such high concentrations is unclear, they may reflect intensive use of DDTs in the 2000s in neighbouring coastal areas because the percentage of DDE relative to tDDT in those samples was close to 50% (Das et al., 2002; Das and Das, 2004), a percentage that implies a relatively recent input into the ecosystem (Aguilar, 1984).

A number of reasons may explain the low concentrations of OCs found in organisms from the Sundarbans. The first and most immediate one is a likely low level of OC inputs into the ecosystem. Bangladesh is one of the poorest and least developed countries in the world. Eighty percent of the population lives in rural areas, industry is weak, and the most important sectors are textiles and clothing, which are not particularly associated with OCs utilization.

A second reason may be the physical and climatic characteristics of the ecosystem. Firstly, the rapid volatilization and high degradation rate of these compounds in tropical environments cause the biological half-lives of semi-volatile compounds, such as DDT, to be shorter (Niimi, 1987). The high temperatures may also increase the rate of elimination of semi-volatile chemicals by fish, due to the influence temperature has on respiratory requirements (Sarkar et al., 2008b). Moreover, the Sundarbans mangrove system is irrigated by three large rivers, the Ganges, the Brahmaputa and the Meghna, which together have formed the largest tropical delta of the planet. To this, it should be added that Bangladesh enjoys the highest rainfall rates in the world and the Sundarbans are moreover subject to large semi-diurnal tides from the Bay of Bengal, with amplitudes of 1-8 m. All this makes it a highly flushed ecosystem, where pollution dilutes and rapidly disperses (Glassby and Roonwal, 1995).

A further reason that appears to contribute to the low pollutant burdens found in the organisms of the Sundarbans is the comparatively short food chain lengths that characterize this ecosystem. As most organochlorine compounds, DDTs and PCBs build up along food webs (*e.g.* Bayen et al., 2005; Reijnders et al., 2018), and the trophic levels determined through the $\delta^{15}N$ values (Cabana and Rasmussen, 1994) of the species analysed in the present study were lower than the reference values given for them in other ecosystems by fishbase.org (Fig. 2), which evidences short food chain lengths. This appears to be an ecological trait of the mangrove ecosystems because similar results have been found in trophic webs of other mangroves (e.g., Ikemoto et al., 2008; Faye et al., 2011; Heithaus et al., 2011). Also, positive relationships between $\delta^{15}N$ values and body length have been observed in a variety of fish species (Overman and Parrish, 2001), a fact that is commonly justified by older individuals feeding at higher trophic levels. Some of the individuals sampled in this study, such as those of *L. calcarifer*, had a body length that corresponded to juvenile individuals (table 1), and this would likely explain the large difference observed between TL $\delta^{15}N$ and that estimated in fisbase.org. Thus, juveniles of this species also consume insects (fishbase org) and they are therefore expected to carry lower OCS concentrations than those typical in a top depredator.

However, other factors may come also into play when interpreting the apparent lack of relationship between trophic levels and OC concentrations. Thus, *S. serrata* displayed higher concentrations of PCB and tDDT than the other species (Fig. 3) despite having a trophic level of only 1.8, but these high concentrations may not be due to biomagnification as crustaceans mostly take up contaminants by diffusion through their body surface or respiratory organs (Gray, 2002; Randall et al., 1998). Indeed, Bonito et al. (2016) already warned that there is no conclusive evidence that OC levels are systematically linked to trophic levels in invertebrate and fish species because bioconcentration rather than biomagnification would be the main process through which organic compounds are accumulated in these organisms (Gray, 2002). This would explain the fact that benthonic species that dwell over muddy bottoms (Table1), *i.e.* the crustaceans *S. serrata* and *P. monodon*, and the fish *M. cephalus, H. nehereus,* and *P. argenteus,* seem to accumulate higher OCs concentrations, probably bioconcentrating them from sediments. All the factors above described, and probably other not identified, likely interplay to produce the observed results.

4.2 Implications for human health of fish and crustaceans consumption

Fresh and saltwater fish are extensively consumed in Bangladesh. However, despite the well-known capacity of fish to accrue OC pollutants and the potential risks that long-term consumption of polluted fish may pose to human health, studies monitoring PCB and tDDT concentrations in edible fish from this area are very limited (Ali et al., 2014). Sinha and Loganathan (2015) reviewed OC concentrations of organisms from different locations of the Ganges River and its tributaries. Concentrations in fish ranged from 20 to 270 ng g^{-1} w.w. for PCBs and from 0.4 to 7,583 ng g^{-1} w.w. for tDDT, and authors took these results as an indication that continued usage of this latter group of compounds in the region facilitated their entering into the Ganges system and, subsequently, into the food web.

Samples of dried fish obtained from markets located in different towns in Bangladesh frequently show large variability in tDDT residues, in some cases reaching relatively high concentrations (Table 4; Bhuiyan et al., 2008, 2009; Hasan et al., 2014). Hasan et al. (2014) measured DDT levels in four commercial species of marine dry fish and found mean concentrations to be lower in production markets than in retail markets. For example, tDDT concentration in samples from the Khulna market were below detection limits except for *P. argenteus* (0.73 ng g^{-1} w.w.), while those from Reajuddin Bazar where much more polluted, with concentrations in *P. argenteus* reaching a level of 228 ng g^{-1} w.w. This difference between locations may not be reflecting environmental pollution, but local use of the pesticide for controlling infestations during processing and storage in some markets (Chowdhury et al., 2010), particularly those whose producers are located far away from consumers (Hasan et al., 2014). Being Khulna market situated at the very fringe of the Sundarbans and thus close to the production areas, the low concentrations found in the present study (*P. argenteus:* 3.31 ng g^{-1} w.w.) as compared with those from other markets (Hasan et al., 2014) are considered to likely reflect true environmental levels of the pesticide.

Table 4. Mean, maximum and minimum levels of tDDT in several species from the Bay of Bengal expressed on a wet weight basis

			tDDT		
Species	**Area**	**mean**	**min.**	**max.**	**References**
Nemipterus japonicus	Northwestern Bay of Bengal	19.9	10.49	30.03	Shailaha and Singbal (1994)
	Madras. Bay of Bengal	0.30			Rajendran et al., (1992)
	Pondicherry. Bay of Bengal	0.16			
	Cuddalore. Bay of Bengal	0.09			
	Tuticorin. Bay of Bengal	0.51			
Sillago spp	Northwestern Bay of Bengal	7.30	3.97	13.88	Shailaha and Singbal (1994)
	Madras. Bay of Bengal	0.05			Rajendran et al., (1992)
	Pondicherry. Bay of Bengal	0.05			
	Cuddalore. Bay of Bengal	0.08			
	Tuticorin. Bay of Bengal	0.07			
Lates Calcarifer	Ganges-Bramhaputra-Meghna estuary	45.06	18.05	72.07	Jabber et al., (2001)
	Madras. Bay of Bengal	0.59			Rajendran et al., (1992)
	Pondicherry. Bay of Bengal	0.05			
	Cuddalore. Bay of Bengal	1.03			
	Tuticorin. Bay of Bengal	2.38			
	Sundarbans	**0.38***	**nd**	**1.29***	**Current study**
Tachysurus thalassinus	South Patches, Bay of Bengal.	468,30	397.09	716,28	Das et al., (2002)
Hilsha ilisha	South Patches, Bay of Bengal	1,965	1,792	2,254	Das and Das (2004)
Pampus argenteus	Madras. Bay of Bengal	0.34			Rajendran et al., (1992)
	Bangladesh city markets (Kulna, Chitagong, Daka)		0.73*	287.64*	Bhuyan et al, 2009; Hassan et al, 2014
	Sundarbans	**3.31***	**nd**	**16.55***	**Current study**
Mugil cephalus	Madras. Bay of Bengal	1.27			Rajendran et al., (1992)
	Tuticorin Bay of Bengal	1.79			
	Sundarbans	**2.05***	**nd**	**5.42***	**Current study**
Harpadon nehereus	Bangladesh city markets (Kulna, Chitagong, Daka)		nd	227.3*	Bhuyan et al, 2008; 2009; Hassan et al, 2014
	Sundarbans	**1.90***	**nd**	**7.99***	**Current study**
Tenualosa ilisha	Bangladesh city markets (Kulna, Chitagong, Daka)		39.61*	72.68*	Bhuyan et al, 2008
	Sundarbans	**2.19***	**0.67**	**4.62***	**Current study**
Peneus monodon	Bangladesh city markets (Kulna, Chitagong, Daka)		nd	152.6*	Bhuyan et al, 2008; 2009; Hassan et al, 2014*
	South-west region of Bangladesh		nd	5	Islam (2017)
	Sundarbans		**nd**	**nd**	**Current study**

* Concentrations converted from d.w to w.w (74% moisture value was used, i.e. d.w *3.85^{-1}).

Tolerance levels for PCBs and DDTs in the edible portion of fish and shellfish have been established at 2,000 and 5,000 ng g^{-1} w.w. respectively (Food and Drug Administration, 2008, 2009). These tolerance levels are approximately 2-3 orders of magnitude higher than the levels found in the fish samples from the Sundarbans and are also much higher that the levels generally reported by other authors in comparable studies in the area (Table 4). Therefore, it is reasonable to conclude that, despite the continued marginal use of pesticides in local agriculture, OC compounds do not pose a sensible risk to the consumer's population through the consumption of fish products. However, this does not mean that enforcement of controls and monitoring are not in order. Rather the contrary, the high OC levels found in dry fish from some retail markets suggest that the potential risk involved in fish consumption outweighs the benefits of using DDT for pest control during fish processing, and point to the need to discontinue such practice and substitute DDT for less hazardous alternatives.

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Figures:

Fig. 1**.** Study area and sampling sites. (1 Khulna market; 2. field). Additionally, the locations of the Ganges-Bramhaputra-Meghna estuary and the South Patches of the Bay of Bengal, where tDDT had previously been measured in fish, are shown.

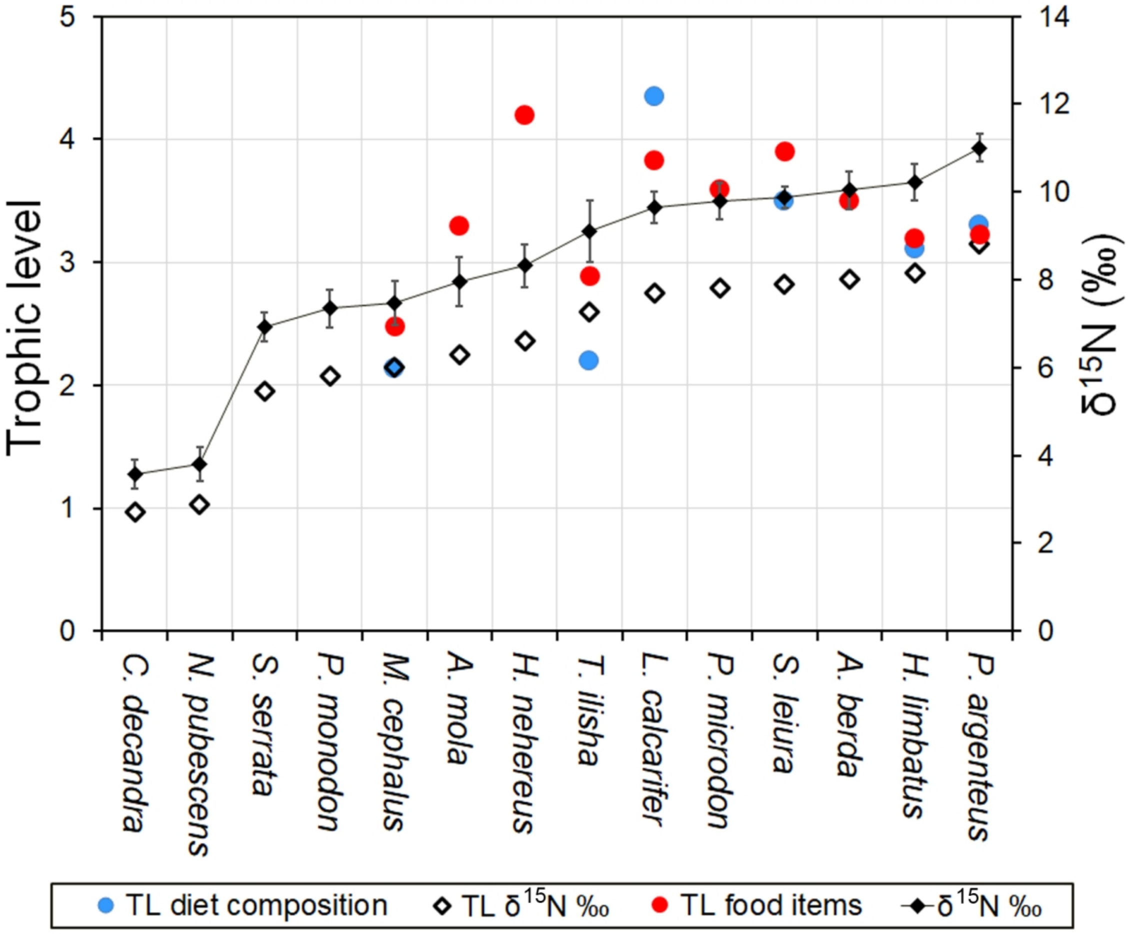

Fig. 2. Mean (±SD) of $\delta^{15}N$ values and trophic levels estimated from 1) $\delta^{15}N$ values, 2) diet composition and 3) a number of food items using a randomized resampling routine (found in fishbase.org)

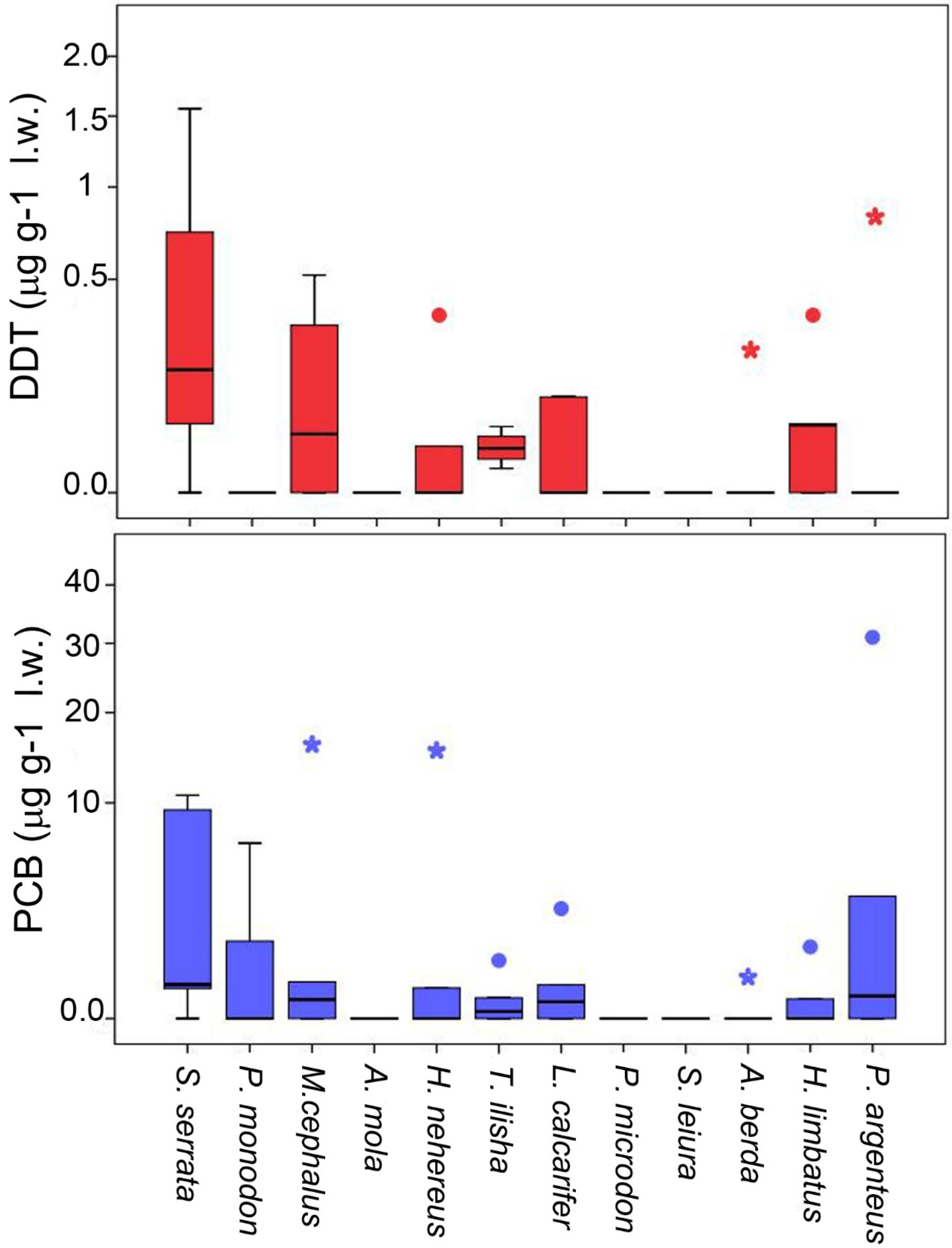

Fig. 3. Boxplot distribution of DDTs and PCB (µg g^{-1} l.w.) for each species. The top and bottom boundaries of each box indicate the 75th and 25th quartile values, respectively, and lines within each box represent the 50th quartile values. The ends of the whiskers indicate the lowest and highest values. Values outside the fence are outliers (○) and extreme outliers (*). Species are ordered according to their trophic level.