Environmental Pollution xxx (2016) 1–9

Contents lists available at ScienceDirect

Environmental Pollution

journal homepage: www.elsevier.com/locate/envpol

Polychlorinated biphenyls and organochlorine pesticides in River Brahmaputra from the outer Himalayan Range and River Hooghly emptying into the Bay of Bengal: Occurrence, sources and ecotoxicological risk assessment☆

Paromita Chakraborty [a, b, *], Sanjenbam Nirmala Khuman [a], Sakthivel Selvaraj [a], Srimurali Sampath [a], Ningombam Linthoingambi Devi [c], John J. Bang [d], Athanasios Katsoyiannis [e]

[a] SRM Research Institute, SRM University, Kattankulathur, Tamil Nadu, India
[b] Department of Civil Engineering, SRM University, Kattankulathur, Tamil Nadu, India
[c] Center for Environmental Sciences, Central University of South Bihar, BIT Campus, Patna 800014, Bihar, India
[d] Department of Pharmaceutical Sciences, North Carolina Central University, USA
[e] Norwegian Institute for Air Research (NILU) at FRAM – High North Research Centre on Climate and the Environment, NO-9296, Tromsø, Norway

ARTICLE INFO

Article history:
Received 26 March 2016
Received in revised form
28 June 2016
Accepted 28 June 2016
Available online xxx

Keywords:
Organochlorine pesticides
Polychlorinated biphenyls
Hooghly
Brahmaputra
Rivers
India

ABSTRACT

River Brahmaputra (RB) from the outer Himalayan Range and River Hooghly (RH), a distributary of River Ganga, are the two largest transboundary perennial rivers supplying freshwater to the northeastern and eastern states of India. Given the history of extensive usage of organochlorine pesticides and increasing industrialization along the banks of these rivers we investigated selected organochlorine pesticides (OCPs) and polychlorinated biphenyls (PCBs) in the surface water of River Brahmaputra and River Hooghly. Geomean of ΣOCPs (53 ng L^{-1}) and Σ_{19}PCBs (108 ng L^{-1}) was higher in RH compared with geomean of ΣOCPs (24 ng L^{-1}) and Σ_{19}PCBs (77 ng L^{-1}) in RB. Among OCPs, γ-HCH showed maximum detection frequency in both the rivers reflecting ongoing lindane usage. DDT and endosulfan residues were observed at specific locations where past or ongoing sources exist. Elevated concentrations of heavier congeners (penta-hepta) were observed in those sites along RH where port and industrial activities were prevalent including informal electronic waste scrap processing units. Furthermore along River Hooghly PCB-126 was high in the suburban industrial belt of Howrah district. PCBs were found to be ubiquitously distributed in RB. Atmospheric transport of tri- and tetra-PCB congeners from the primary source regions might be a major contributor for PCBs in RB. Heavier congeners (penta-nona) in the urban centers of RB were likely due to industrial wastewater runoff from the oil refineries in the Brahmaputra valley. Σ_{19}PCBs concentrations in this study exceeded the USEPA recommended limit for freshwater. Ecotoxicological risk assessment showed the possibility of adverse impact on the organisms in the lower trophic level due to DDT and lindane contamination. Impact of endosulfan on fishes might be of considerable concern for aquatic environment.

1. Introduction

Freshwater is the basic need for human survival and the inherent ecosystems support diverse group of flora and fauna. India has 16% of the global population with only 4% of the world's freshwater. Currently the freshwater quality in India and associated biodiversity are facing serious challenges due to the fast growing economy, increasing industrialization and the large agricultural sector. River Brahmaputra (RB) is a trans-boundary river that enters India in Arunachal Pradesh along the outer Himalayan Range and flows from Assam to West Bengal where it meets River Hooghly (RH), a distributary of River Ganga. RB and RH merges with River Meghna in Bangladesh and before emptying into the Bay of Bengal forms the world's largest delta, the Sunderban delta shared by the

☆ This paper has been recommended for acceptance by Chen Da.

* Corresponding author.SRM Research Institute, SRM University, Kattankulathur, Tamil Nadu, India.
E-mail address: paromita.c@res.srmuniv.ac.in (P. Chakraborty).

http://dx.doi.org/10.1016/j.envpol.2016.06.067

state of West Bengal in India and Bangladesh. RB and RH are two perennial rivers providing perpetual supply of freshwater to the plains of eastern and northeastern states of India to support irrigation, human and industrial consumption. From River Hooghly, nearly 493 million gallons per day is pumped to satisfy water demand in the surrounding region (Rudra, 2007). Central pollution control board reported that 1573 million litres per day (MLD) of sewage is discharged in RH (CPCB, 2008). Brahmaputra basin has a population of 29.1 million people utilizing 24 km^3 of surface water every year (Sandrp, 2013).

During 2005–2010, 5039 metric tons (MT) and 1191 MT of dichlorodiphenyl trichloroethane (DDT), were used for malaria control in Assam and West Bengal respectively and 17856 MT of endosulfan was produced in India (PPIN, 2013). Synthetic pesticides constituted 85% of the total pesticide demand in the northeastern India and one fourth of those pesticides were organochlorine pesticides (OCPs). Among OCPs, endosulfan was mostly applied in the tea estates (Gurusubramanian et al., 2008). Aldrin and dieldrin were never produced in India but 622 MT of aldrin was imported during 1995–2000 (GEFIO, 2002).

The inventory of electrical equipment containing PCBs estimated 1548 transformers containing pure and contaminated PCBs with total weight of PCB oils to be 10000 MT and the total amount of PCBs is estimated to be 28000 MT in India (Sharma, 2013).

In the recent past, atmospheric OCPs (Chakraborty et al., 2010, 2013) and PCBs (Chakraborty et al., 2013) have been observed in and around Kolkata city along the River Hooghly and the authors of those studies also reported associated re-emission from soil (Chakraborty et al., 2015, 2016). Hooghly estuarine sediment has been found to be contaminated with organic contaminants (Bhattacharya et al., 2003; Guzzella et al., 2005). OCPs may reach the surface riverine water via agricultural runoff, direct applications, spray drift, aerial spraying and erosion (Turgut, 2003; Singh et al., 2007; Zhou et al., 2008).

Soil borne OCP residues have been reported from the northeastern states of India along the bank of River Brahmaputra (Mishra et al., 2012; Devi and Raha, 2013). Persistent organic pollutants (POPs) like OCPs and PCBs owing to their resistance to degradation, toxicity, bioaccumulation through food chain and their ability for long range atmospheric transport (Erdogrul et al., 2005; Guzzella et al., 2005; Gao et al., 2008) are of serious concern for freshwater quality.

Though India is a signatory for the Stockholm convention (Fiedler, 2008), only limited data is available for such compounds from the surface water of perennial rivers, particularly from eastern and northeastern states of India. Hence in this study we aim to elucidate the occurrence and sources of selected OCPs and PCBs in the surface water of River Brahmaputra from the outer Himalayan Range and lower stretch of River Hooghly before emptying into the Bay of Bengal and estimate the associated risk posed to the inherent freshwater ecosystems.

2. Materials and methods

2.1. Study area

Details of the sampling sites have been given in Fig. 1, Table S1 and S2. Surface river water samples were collected using a stainless steel bucket from 16 sites in RB and 20 sites in RH during summer in the month of June 2012 (Fig. 1a and b). For each site, a composite water sample was collected over a length of 500 m from 5 points. Five samples within nearly 9 m^2 of each site (each of 1 L) of river water were collected and mixed well to obtain one composite sample for each site. Sampling jars were pre-washed with organic solvent (methanol) before sampling. River water samples for chemical analysis were stored in pre-cleaned amber, glass bottles and collected in triplicates. The triplicate samples were used for backup purposes (in case of breakage of the primary sample) and for laboratory replicates. Following collection, samples were stored in an ice box and sent to the laboratory on the same evening and kept at 4 °C until analysis.

2.2. Extraction and analysis

OCPs and PCBs were extracted using liquid–liquid extraction method. Approximately one litre of unfiltered water sample (triplicate from each site) was transferred into a separatory funnel and 10 g of sodium chloride (NaCl) was added and shaken well to completely dissolve NaCl. Known amounts of 2,4,5,6-tetrachloro-m-xylene (TCmX) and PCB-209 (decachlorobiphenyl) were added in each sample as recovery surrogates. After adding dichloromethane (DCM) thrice (50, 25, 25 mL), each sample was shaken vigorously for 3–5 mins for 3 times. The organic phase was collected and dried by passing through 3 cm of sodium sulfate placed on glass wool. The extract was concentrated and transferred into *n*-hexane and further purified on a 8 mm i.d. alumina/silica column packed with neutral alumina (6 cm, 3% deactivated), neutral silica gel (10 cm, 3% deactivated), 50% (w/w) sulfuric acid silica (10 cm), and anhydrous sodium sulfate (1 cm). Before use, neutral alumina, neutral silica gel, and anhydrous sodium sulfate were Soxhlet extracted for 48 h with dichloromethane (DCM), and baked for purification. The column was eluted with 50 ml of DCM/hexane (1: 1, v/v) to yield the OCPs and PCBs fraction. The obtained sample extracts were allowed to pass through anhydrous sodium sulfate to remove traces of water contents and then concentrated by means of rotary evaporation (Buchi, Switzerland) under reduced pressure at 40 °C to 1 mL. PCB-54 was added as internal standard prior to analysis. Finally 1 μL was injected into GC-MS. The chromatographic separation and quantification of pesticides were carried out using Agilent Gas Chromatograph (7890B) equipped with Mass spectrometer (5977A). Separation of target compounds was performed by using CP-Sil 8 CB capillary column (50 m × 0.25 mm i.d. × 0.25 μm film thickness) operating under selected ion monitoring mode. The column temperature was programmed up to 120 °C for 1 min then increased with 3 °C min^{-1} to 210 °C, kept for 1 min, then further ramped by 10 °C min^{-1} to 290 °C and kept for 5 mins. The injector and detector temperatures were 225 °C and 300 °C, respectively. Purified Helium was used as a carrier gas at the flow rate of 1.0 mL min^{-1}. 19 PCB congeners (PCB-28, -37, -44, -52, -70, -74, -81, -118, -119, -123, -126, -138, -151, -156, -161, -168, -177, -189 and -207) in RB and 19 PCB congeners in RH (PCB-28, -37, -44, -52, -70, -74, -114, -118, -119, -123, -126, -138, -151, -167 -168, -169, -177, -189 and -207) and 13 OCPs (α-, β-, γ- and δ-HCH, *o,p*′-DDT, *p,p*′-DDE, *p,p*′-DDT, and *p,p*′-DDD, heptachlor, aldrin, dieldrin, and α- and β-endosulfan) were regularly detected in samples and quantified.

2.3. Quality assurance/quality control

Requisite quality assurance and quality control (QA/QC) was performed, including procedural blanks (analyte concentrations were < method detection limit, MDL), random duplicate samples, five level calibration curves ($R^2 = 0.9998$), calibration verification, and surrogate recovery. Each sample was analyzed in duplicate and the average was used in the subsequent calculations. The instrument was calibrated with every batch of sample analysis. The instrument detection limits with valid quantifiable peak were calculated using signal to noise ratio >3:1. Method detection limit (MDL) ranged between 0.019 and 0.082 ng L^{-1} for PCBs and 0.009 and 0.01 ng L^{-1} for OCPs, respectively. Surrogate recovery for TCmX

Please cite this article in press as: Chakraborty, P., et al., Polychlorinated biphenyls and organochlorine pesticides in River Brahmaputra from the outer Himalayan Range and River Hooghly emptying into the Bay of Bengal: Occurrence, sources and ecotoxicological risk assessment, Environmental Pollution (2016), http://dx.doi.org/10.1016/j.envpol.2016.06.067

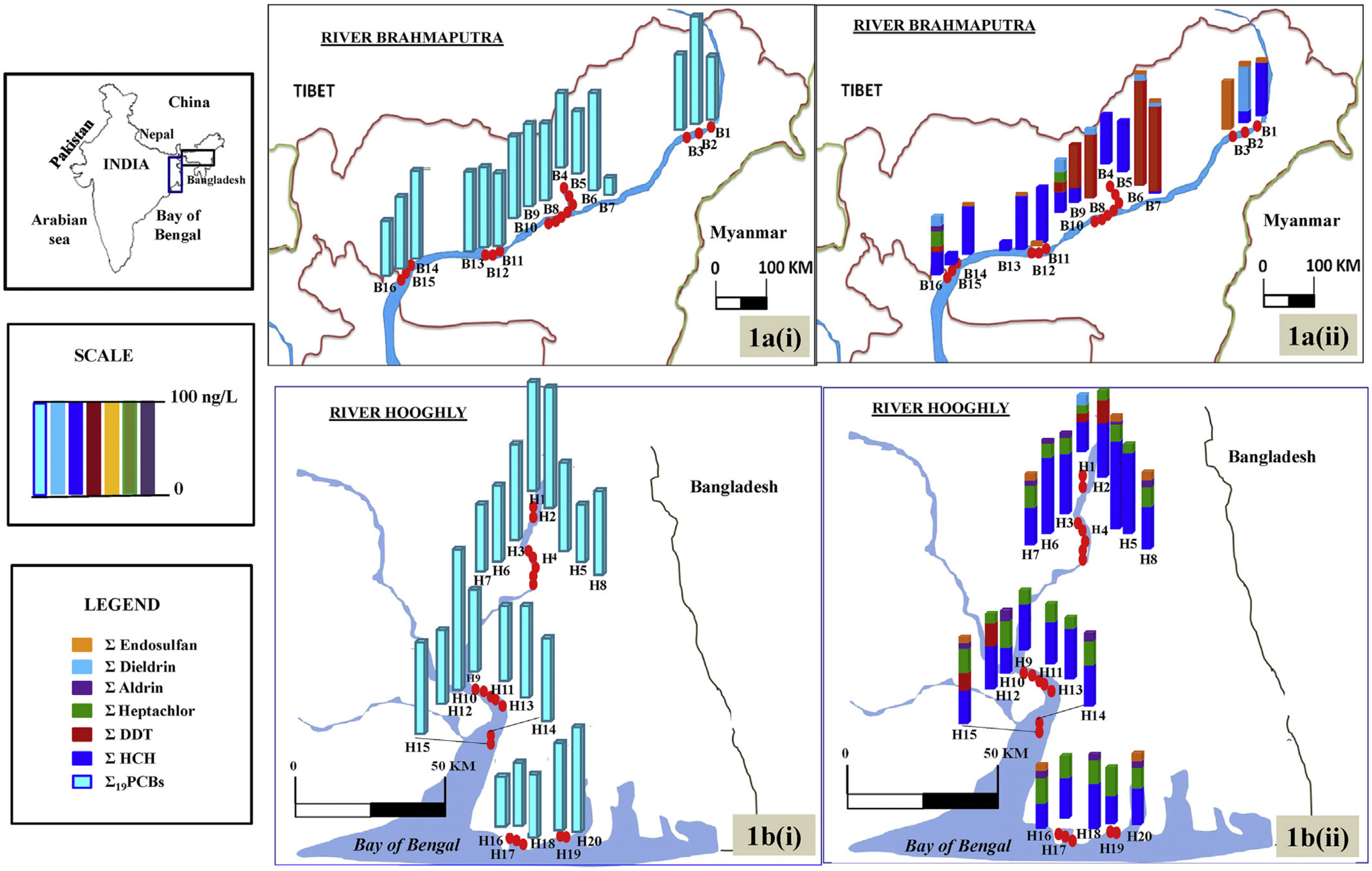

Fig. 1. a. Sampling sites in River Brahmaputra and distribution of (i) Σ_{19}PCBs and (ii) ΣOCPs; b. Sampling sites in River Hooghly and distribution of (i) Σ_{19}PCBs and (ii) ΣOCPs.

and PCB-209 ranged between 91 and 118%.

2.4. Principal component analysis (PCA)

PCA is multivariate analysis widely used to identify/track the possible sources. Moreover, it is a technique to bring out a strong model and highlight variation of datasets. It has been used to identify the sources for PCB congeners in RB and RH. Further, KMO, coefficients, determinant and Bartlett's test of sphericity were applied to extract dataset into three factors with maximum of 25 iterations for convergence. Varimax rotation was used and factors scores were saved as regression variables. PCA was done in SPSS version 19.

2.5. Pollution assessment

Risk assessment was estimated in terms of risk quotient as given elsewhere (USEPA, 2006). Details of this methodology has been given in Supporting Information.

Further, ecotoxicological risk assessment has been estimated for DDT, endosulfan and lindane by calculating the hazard quotient (HQ) based on USEPA guidelines (USEPA, 1998).

Precisely, HQ has been estimated using the given formula:

$$HQ = \mathrm{MEC}/\mathrm{PNEC} \quad (1)$$

where, MEC is the maximum reported environmental concentration i.e. the maximum concentration found in the surface riverine water and PNEC is the predicted no-effect concentration of aquatic species. PNEC values for five species based on the available toxic end point value in five groups along different trophic levels viz., zooplankton, phytoplankton, molluscs, insects, fishes were calculated by dividing acute toxicity values by a factor of 1000 and chronic toxicity values by a factor of 10 as per calculation given elsewhere (Ramaswamy et al., 2011; Selvaraj et al., 2014). Type of organism, toxicity endpoints and PNEC values are given in Tables S3, S4 &S5. HQ exceeding 1 indicates that the organism is vulnerable (affected) for the given concentration of environmental toxicant.

2.6. Statistical analysis

All statistical analyses were performed using SPSS version 19.

3. Results and discussion

3.1. General discussion

Overall concentration range and spatial distribution of $\sum_{19}$PCBs and selected OCPs in River Brahmaputra (RB) and River Hooghly (RH) have been given in Table 1 and Fig. 1a & b. $\sum_{19}$PCBs varied between 39–161 ng L^{-1} (average ± SD: 83 ng L^{-1} ± 32) in RB and 57–233 ng L^{-1} (average ± SD: 116 ng L^{-1} ± 46) in RH. More than 90% PCBs were comprised of tri and tetra congeners in both the rivers in line with Pearl River, China (Guan et al., 2009). Dominance of tetra congeners were also observed in River Yamuna in New Delhi, India (Kumar et al., 2012, 2013) and River Chenab in Pakistan (Eqani et al., 2012). Similarly lighter chlorinated biphenyls (di-, tri- and tetra-) were found to be high in the surface water of Sao Paulo River, Brazil (Rissato et al., 2006).

Table 1
Concentrations of selected organochlorine pesticides and polychlorinated biphenyls in River Brahmaputra and River Hooghly.

	River Brahmaputra (n = 16)		River Hooghly (n = 20)	
Compounds	Range (in ngL^{-1})	Avg ± stdev	Range (in ngL^{-1})	Avg ± stdev
α-HCH	ND – 6	1 ± 2	ND –22	3 ± 5
β-HCH	ND – 6	0.4 ± 2	ND – 16	3 ± 6
γ-HCH	ND – 14	6 ± 7	ND – 28	13 ± 8
δ-HCH	ND – 12	1 ± 3	ND– 114	19 ± 33
ΣHCHs	ND – 22	8 ± 8	8–114	3 ± 28
p,p′-DDD	ND – 113	14 ± 33	ND	ND
o,p′-DDT	ND – 113	15 ± 34	ND – 26	3 ± 8
p,p′-DDT	ND – 7	1 ± 2	ND – 5	0.3 ± 1
ΣDDTs	ND – 225	30 ± 66	ND – 26	4 ± 8
Heptachlor	ND – 10	1 ± 3	5–26	14 ± 6
Aldrin	ND – 5	0.3 ± 1	ND – 9	4 ± 4
Dieldrin	ND – 19	4 ± 6	ND – 7	0.4 ± 2
ΣAldrin	ND – 19	4 ± 6	ND – 9	4 ± 4
α-Endosulfan	ND – 9	1 ± 2	ND	ND
β-Endosulfan	ND – 45	3 ± 11	ND – 10	2 ± 4
ΣEndos	ND – 53	3 ± 13	ND – 10	2 ± 4
ΣOCPs	2 – 245	47 ± 67	12–154	62 ± 35
$\sum_{19}$PCBs	39–161	83 ± 32	57–233	116 ± 46
∑dl-PCBs	ND – 2	1 ± 1	ND – 5	2 ± 2

ΣOCPs ranged from 2 to 245 ng L^{-1} (average ± SD: 47 ng L^{-1} ± 67) and 12–154 ng L^{-1} (average ± SD: 62 ng L^{-1} ± 35) in RB and RH respectively. High detection frequency for OCPs was observed in both RB (88%) and RH (100%). Paired T-test did not show any significant difference in the distribution of OCPs in these two rivers.

When compared with Indian rivers, HCHs, DDTs, endosulfan and heptachlor contamination in the surface water of RB and RH were lower than the upper stretches of River Ganga (Singh et al., 2007), other tributaries of River Ganga and River Gomti in Uttar Pradesh (Malik et al., 2009), River Yamuna in Haryana and Delhi (Kaushik et al., 2008) and River Ghaggar in Haryana (Kaushik et al., 2010). Relatively higher pesticidal usage for agriculture in the vast Indo-Gangetic plains of northern India compared to eastern and north-eastern states is the most plausible reason for the current observation (Table S6).

DDT concentrations in RH were comparable to surface water of Yangtze River (Sun et al., 2002) and Qiantang River (Zhou et al., 2008) in China. Notwithstanding the fact that POPs globally declined (AMAP, 2014), the present POPs concentrations have been found to be higher than what was detected in rivers a decade ago in other parts of the world like Turkey (Turgut, 2003), South Africa (Sibali et al., 2008) and recently in River Chenab from Pakistan (Eqani et al., 2012) but in line with the OCP concentrations in Densu River basin, Ghana (Kuranchie-Mensah et al., 2012). HCHs in RH was comparable with River Chenab in Pakistan (Eqani et al., 2012), but higher than rivers in China (Na et al., 2006; Zhou et al., 2008) and much higher than the Arctic region (Webster, 2004). However, the concentration of HCHs in this study was lower than River Kucuk Menderes, Turkey (Turgut, 2003) and Jukskei River catchment area in Gauteng, South Africa (Sibali et al., 2008).

3.2. Distribution and sources

3.2.1. PCBs

Highest concentration of $\sum_{19}$PCBs was observed in Hooghly at Namkhana Port (site H10, 233 ng L^{-1}) followed by Belurmath (site H6, 194 ng L^{-1}) and Chinsura (site H2, 186 ng L^{-1}). PCB-28, -52 and -118 were detected in all the sites of RB and RH. Among the heavier congeners only PCB-177 was found in specific locations of RB (B8, B9, B10, B12, B15, B16) and RH (H3, H4, H12 and H15), possibly indicating localized sources. Details of composition of PCB congeners for RH and RB have been given in Fig. 2a and Fig. 3a respectively.

Three dimensional PCA for detected congeners having more than 60% detection frequency in RB and RH having best fit (>0.6) were used to identify PCB sources (Figs. 2b and 3b, Table S9). Along RH, PCB congeners in PC-1 were PCB-44, -52, -169, -189, -207; PC-2 congeners were PCB-28, -123, -151 and PC-3 congeners were PCB-168, -126, -118 with variance of 22%, 20% and 16% for PC-1, PC-2 and PC-3 respectively. In RH, tetra congeners in PC-1, PCB-44 and PCB-52 contributed 53% of the total PCB concentration (Fig. 2b). Dominance of tetra congeners in surface soil and atmospheric emission of tetra congeners particularly during open burning of municipal solid waste has been reported from Kolkata (Chakraborty et al., 2016). Atmospheric deposition might be a possible reason for these congeners in River Hooghly. Moreover electronic scraps are informally recycled in the suburban industrial belt of Howrah district along River Hooghly. Further some amount of electronic wastes also end up in open dumpsites (Chakraborty et al., 2016). First eight sites of RH (H1–H8, Fig. 1) located in the suburbs near Kolkata are surrounded by plenty of small and large scale industries. Large scale scrap processors dealing in electronic waste scrap from multiple industries are present in this part of Howrah district. We observed elevated concentration of total PCBs in these eight sites (Geomean, 122 ng L^{-1}) with higher percentage of tetra congeners (Fig. 2a). In addition to tetra congeners, heavier congeners like PCB-169, -189 and -207 were prevalent in these sites. Hence we suspect industrial wastes runoff particularly during monsoonal rain might be another possible source for PCBs in RH.

PCB-28 contributed 40% of the total PCB concentration. Maximum PCB-28 was observed at Namkhana port (H-10, 142 ng L^{-1}) possibly associated with port activities (Chakraborty et al., 2016). Further heavier congeners like PCB-123 and -151 were found in 12 sites (Site H9 to H20; Geomean, 100 ng L^{-1}) adjacent to Namkhana port up to the tip of the Bay of Bengal (Fig. 2a). Among the two major ports of eastern India namely Kolkata and Haldia, the former is situated within the Hooghly estuary. These are the only riverine ports of India engaged in container handling after Vishakhapatnam, located in southern part of India. Harbor or port activity is likely acting as a source for PCBs in the lowermost stretch of RH close to the Bay of Bengal. PCB contamination due to harbor activities in southern India along the Bay of Bengal has been observed earlier (Rajendran et al., 2005). Further transboundary movement of PCBs from the ship breaking sites of Bangladesh (Nøst et al., 2015) might be another possible source of PCBs in the lower stretch of RH.

Highest PCB-118 was detected at H2. PCB-168 was observed only at H1, H2 and H3 with concentration ranging between 1 to 2 ng L^{-1}. The most toxic congener, PCB 126, was detected in six out of the eight sites in the suburban industrial belt and rural pockets. In the rural sites (H1 and H2) and suburban sites (H4, H6, H8 and H9), PCB-126 could be due to discharge of combusted coal and industrial wastes (Chi et al., 2007).

In River Brahmaputra PCB-28, -44, -74, -119, -151 and -169 were strongly weighted in PC-1 with 29% variance. PCB-28 and -44 were ubiquitously distributed in RB and contributed 29% of total PCB concentration. Percentage of tri-CB was higher in RB compared with RH (Fig. 2a and 3a). We found that the samples taken from the distributary of River Brahmaputra from Arunachal Pradesh (B4–B10) exhibited consistently elevated PCBs (mean of 77 ng L^{-1}). In the recent past high PCBs in Manipur (Devi et al., 2014), a northeastern state of India, was attributed to long range atmospheric transport of PCBs from source regions in Kolkata (Chakraborty et al., 2013). Monsoon-driven long range atmospheric transport of PCBs in the high altitude and cold climate of the outer

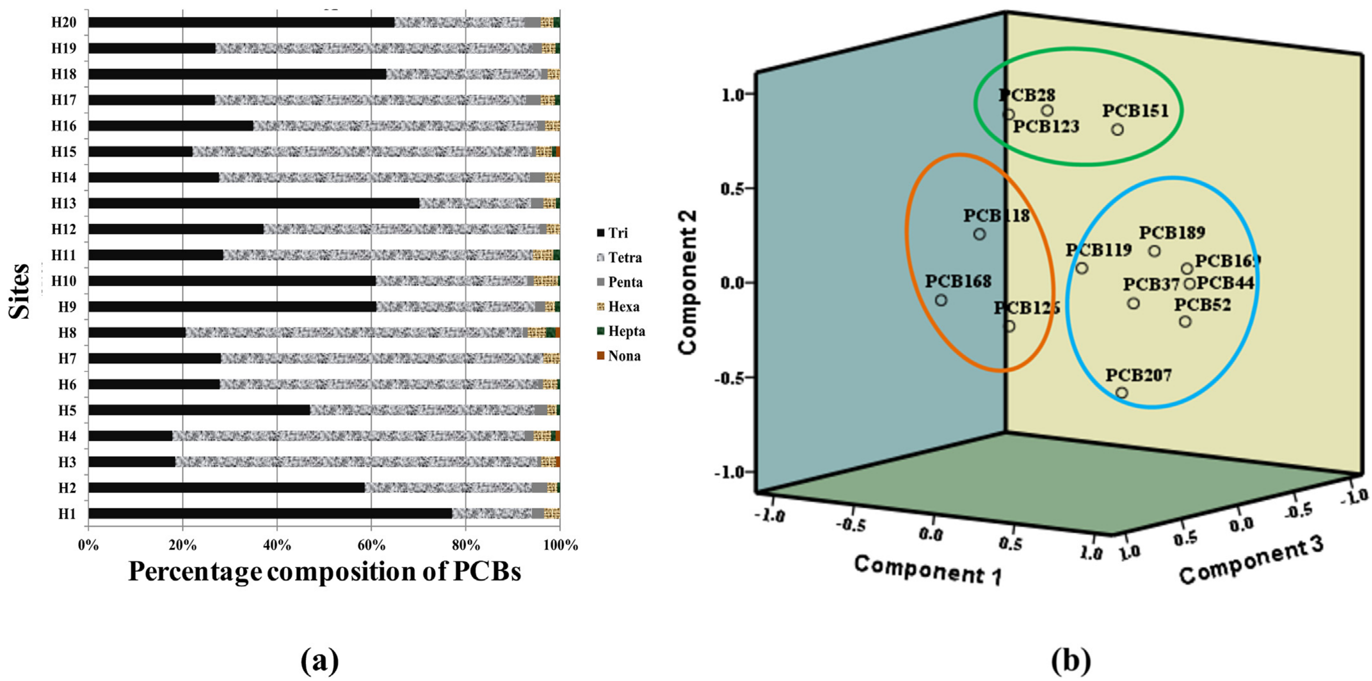

Fig. 2. (a). Percentage composition and (b). Source of PCB congeners using PCA in River Hooghly.

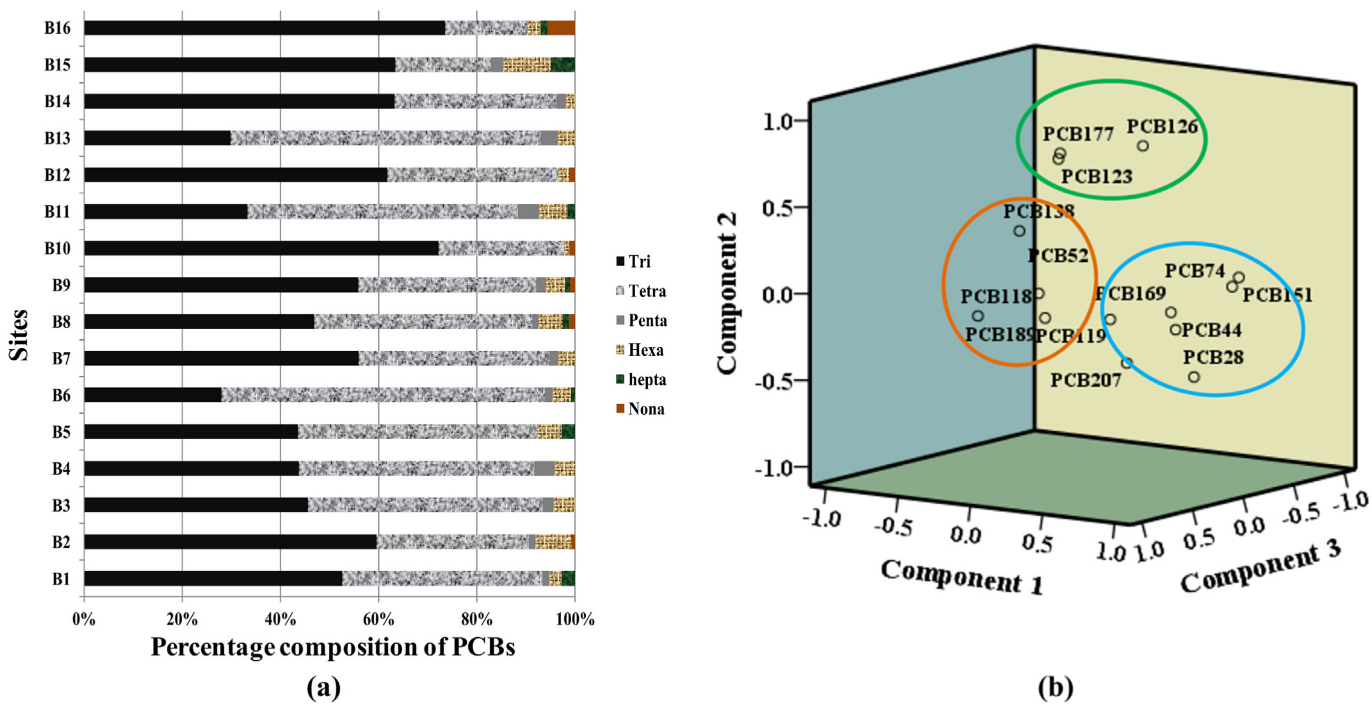

Fig. 3. (a) Percentage composition and (b) Sources of PCB congeners using PCA in River Brahmaputra.

Himalayan range has been observed earlier (Kang et al., 2009; Sheng et al., 2013). Hence we speculate that the dominance of lighter PCB congeners in all the sites of RB might have resulted from atmospheric deposition (Semeena et al., 2006). Lower concentration of medium and high molecular weight congeners like PCB-119, -151, -169 might be associated with sludge or wastewater runoff.

Among PCB- 126, -123 and -177 in PC-2, PCB -126 was observed in more than 80% sites of RB. Assam is a major producer of crude oil and natural gas in India with the second oldest oil well in the world. Most of the oilfields of Assam are located in the Upper Assam region of the Brahmaputra Valley. Apart from PCB-126, heavier congeners like PCB-123 and -177 also significantly contributed for this component. We suspect that in RB, the riverine discharge during combustion of coal and industrial waste may be the major contributor for PCB-126 (Chi et al., 2007).

PCB -52 and -118 were loaded in PC-3. Like RH, PCB-52 was

Please cite this article in press as: Chakraborty, P., et al., Polychlorinated biphenyls and organochlorine pesticides in River Brahmaputra from the outer Himalayan Range and River Hooghly emptying into the Bay of Bengal: Occurrence, sources and ecotoxicological risk assessment, Environmental Pollution (2016), http://dx.doi.org/10.1016/j.envpol.2016.06.067

found in all the sites of RB presumably for the same reason as that of RH i.e., discharge from open burning of dumped solid waste. Direct discharge of ashes including plastics and wood especially in the burning ghats along the bank of these rivers were found to pollute river water. PCB-118 contributed more than 30% of dl-PCBs indicating municipal solid waste incineration (MSWI) pattern (Liu et al., 2013).

3.2.2. OCPs

OCPs were found in the following order: DDTs > HCHs > Dieldrin > Endosulfan > Heptaclor > Aldrin in RB. In RH the order of OCPs was: HCHs > Heptachlor > Aldrin > DDTs > Endosulfan > Dieldrin. HCHs concentrations were relatively high in RH (Geomean, 31 ng L^{-1}) and ΣHCHs was significantly correlated with ΣOCPs ($R^2 = 0.989$, $p < 0.01$). Highest DDT concentration was observed in RB (Geomean 49 ng L^{-1}) and DDTs showed significant correlation with ΣOCPs ($R^2 = 0.976$, $p < 0.01$). Significant correlations were observed between ΣOCPs and concentration of heptachlor ($R^2 = 0.920$, $p < 0.01$) and endosulfan ($R^2 = 0.502$, $p < 0.05$) in RH. Extensive historical usage of OCPs along the bank of River Hooghly for agricultural purpose might have resulted in such a correlation.

3.3. HCHs

ΣHCHs ranged between ND (not detected) −22 ng L^{-1} and 8−114 ng L^{-1} in RB and RH respectively. Overall γ-HCH dominated in both the Rivers accounting for more than 50% of ΣHCHs. Highest concentrations of ∑HCHs were found in the suburban sites of Howrah district (H3, 114 ng L^{-1} and H6, 83 ng L^{-1}). Earlier studies reported elevated levels of HCHs in Hooghly estuarine sediment (Bhattacharya et al., 2003). High concentration of HCHs have been observed both in air (Chakraborty et al., 2010) and soil (Chakraborty et al., 2015) in and around Kolkata. In RH, 35% of the sites showed β/(α + γ) isomeric ratios less than 1 suggesting ongoing usage of lindane. Whereas, in two sites of RH (H12 & H17) and in three sites of RB (B1, B13 and B16), α-HCH was predominant thereby indicating the ongoing usage of technical HCH. High concentration of δ-HCH in 60% sites was similar to the observation in Gomti River, India (Malik et al., 2009).

3.4. DDTs

ΣDDTs varied between ND − 225 ng L^{-1} in RB and ND − 26 ng L^{-1} in RH. In RB, *p,p*′-DDD was predominant (ND − 113 ng L^{-1}; average ± SD:13 ± 31 ng L^{-1}). High *o,p*′-DDT (ND − 113 ng L^{-1}, average = 13 ng L^{-1}) and to some extent *p,p*′-DDT (ND − 7 ng L^{-1}; average = 1 ng L^{-1}) can be attributed to the ongoing usage of DDT for vector control in northeastern India (Devi et al., 2011; Mishra et al., 2012; Devi and Raha, 2013). During 2006−2007, India used 6000 and 2560 MT of DDT for control of Malaria and Kalazar respectively (Gupta, 2004). Among all the Indian states Assam consumed the highest DDT for vector control during 2005−2010 (NVBDCP, 2010).

In RH *o,p*′-DDT was higher than *p,p*′ DDT. Presence of parent isomers like *o,p*′-DDT (ND − 26 ng L^{-1}) and *p,p*′-DDT (ND − 5 ng L^{-1}), which constitute 90% of the technical DDT (*p,p*′-DDT = 75%, *o,p*′-DDT = 15%) is another indication that fresh DDT usage take place, possibly for vector control in RH, something that is consistent with observation in different environmental matrices from this region (Chakraborty et al., 2010, 2014, 2015).

3.4.1. Endosulfan

Endosulfan is a versatile broad spectrum pesticide which was extensively used in India. In 2007 consumption of endosulfan in India recorded a total of 12 million litres out of which West Bengal and Sikkim consumed 1.1 million litres and Assam and other northeastern state consumed 0.9 million litres (Guide, 2011).

Maximum endosulfan concentration was found in RB from upper Assam (site B1, 53 ng L^{-1}). In this site β-endosulfan is five-folds higher than α-endosulfan. For nearly 60% sites in RB, both α- and β-isomers were detected. Relatively high concentrations of α-endosulfan and β-endosulfan were found in Dibrugarh (B1 to B3), a region well known for tea plantations. For more than 100 years endosulfan has been used for pest control in the tea estates of northeastern India and high levels of endosulfan residues were recovered in tea from this region (Gurusubramanian et al., 2008). Recently high levels of endosulfan was reported in soil from northeastern states of India particularly from Assam (Mishra et al., 2012; Devi and Raha, 2013).

Only β-endosulfan was detected in six sites of RH ranging between ND − 10 ng L^{-1} with the highest concentration at site H4. Higher detection frequency of β-endosulfan in RH could be associated with historic usage of endosulfan along the bank of Hooghly thereby showing elevated concentrations in soil (Chakraborty et al., 2015), atmosphere (Chakraborty et al., 2010) and estuarine sediment (Guzzella et al., 2005). Current observation can be reasoned with the fact that the water solubility of β-endosulfan is ten folds higher than α-endosulfan. Moreover β-endosulfan is likely to undergo higher atmospheric deposition than α-endosulfan (Weber et al., 2010).

3.4.2. Other pesticides

Heptachlor is primarily used against soil insects and termites. It has been used against cotton insects, grasshoppers, some crop pests and to combat malaria. Heptachlor was observed in all the sites of RH, and varied between 5 and 26 ng L^{-1} (average ± SD, 14 ± 6.5 ng L^{-1}). In RB heptachlor ranged between ND − 10 ng L^{-1} (average ± SD, 2 ± 4.3 ng L^{-1}). Highest concentration of heptachlor was found in RH (site H15). Heptachlor was detected only in two sites of RB possibly associated with the localized heptachlor usage in Assam.

Aldrin was used as anti-termite agent on potato crops before being banned in 1996 (Devi and Raha, 2013). West Bengal is the second largest producer of potato in the country and produced 11.5 million tonnes during 2012−2013 (Agriculture, 2013). In RH aldrin concentration was slightly higher (ND − 9 ngL^{-1}) than dieldrin (ND − 7 ngL^{-1}) possibly suggesting fresher input. In RB, dieldrin was found to be higher (ND − 19 ng L^{-1}) than aldrin (ND − 5 ng L^{-1}). Highest concentration of dieldrin was detected at site B3 in RB. Higher dieldrin concentration might be due to lower volatility and conversion of aldrin to dieldrin in environment (Jorgenson, 2001). Hence dieldrin in RB is likely from an aged source.

3.5. Pollution assessment

OCPs and PCBs concentrations were compared with the current EPA recommended national water quality criteria (USEPA, 2006) (Table S11 and S12). Risk Quotients for criteria continuous concentration (RQ_{CCCs}) exceeded the limit ($RQ_{CCCs} > 10$) in majority sites of both the rivers for OCPs and PCBs and the observations are in line with River Chenab in Pakistan (Eqani et al., 2012). PCB occurrence in RB and RH is comparable or higher than the raw water standard of developing countries like China (ChinaSEPA, 2002) (<20 ng L^{-1}). Toxicity equivalents (TEQs) associated with dl-PCBs were almost similar for both RB (0.01−0.17 ng TEQ/L) and RH (0.02−0.2 ng TEQ/L) (Table S10). PCB-126 dominated the TEQ particularly in the urban centers along RB and close to the suburban industrial region and sites having port activities along RH.

Please cite this article in press as: Chakraborty, P., et al., Polychlorinated biphenyls and organochlorine pesticides in River Brahmaputra from the outer Himalayan Range and River Hooghly emptying into the Bay of Bengal: Occurrence, sources and ecotoxicological risk assessment, Environmental Pollution (2016), http://dx.doi.org/10.1016/j.envpol.2016.06.067

3.6. Ecotoxicological concern

Ecotoxicological risks for flora and fauna for those sites detected with OCP residues have been shown in the form of boxplots (Fig. 4) using the hazard quotient (HQ) approach (Selvaraj et al., 2014). Generally, we observed that in the lower trophic levels phytoplanktons like algae, dinoflagellate and diatom and zooplankton like waterflea, scud and grass shrimps were affected by lindane, DDT and endosulfan in both the rivers. The percentage of species affected in each group has been given in Supporting Information (Fig. S1). Currently the impact of elevated DDT and lindane concentration was limited to lower trophic level (Phytoplankton and Zooplankton) therefore the biodiversity in these freshwater ecosystems were likely to be at risk. Moving to the higher trophic level, insects like Mayfly were affected by DDT, endosulfan and lindane. Maximum impact of DDT on mosquitoes in RB can be attributed to the DDT usage for vector control in this region. Unlike DDT and lindane, even with lesser surface water concentration endosulfan showed maximum impact on fishes. Endosulfan is known to be highly toxic to aquatic species and fishes are extremely sensitive to endosulfan (Fan, 2007). In this study endosulfan showed higher impact on fishes particularly on four edible fish species viz., *Catla catla*, *Anguilla anguilla*, *Lepomis macrochirus and Labeo rohita* in RB and two species in RH (*Catla catla* and *Lepomis macrochirus*). West Bengal is the leading inland fish and fish seed producer in the country contributing 11.7 lakh tonnes of fish to the national food basket (ICAR, 2016). Fishes form an integral part of the regular diet for majority people in the eastern and northeastern states of India particularly *Catla catla* is one of the major carp available in this region. Typical fish flesh consumption among residents of West Bengal, particularly in the middle income groups, ranges between 300 g to 500 g per week per person (Kumar, 2010). Apart from possible risk on aquatic environment, endosulfan being hydrophobic, highly lipophilic with potential to bioconcentrate and biomagnify in food chain might be also a concern for human health. However, an in-depth study is required to understand the direct pathway for bioaccumulation of such organochlorine compounds in this region.

3.7. Conclusion

Extensive usage of OCPs in the past and limited ongoing usage resulted in surface riverine water contamination of OCPs in River Brahmaputra and River Hooghly. Atmospheric deposition and agricultural run-off are the potential pathways for OCPs in these two transboundary perennial rivers. Apart from DDT usage for vector control, fresh inputs of other OCPs, particularly endosulfan, in certain pockets of eastern and northeastern states of India seems to be prevalent. Unlike OCPs, PCBs were ubiquitiously distributed in both the rivers with elevated concentration in Hooghly particularly in those pockets where industrial and port activities were prevalent along the bank of the river. We suspect that in the outer Himalayan range lighter PCB congeners might have been influenced by long range atmospheric transport from the source regions. PCBs concentrations in all the sites were above the safety limits for aquatic organisms which is an indication of probable risk for such organisms. Estimated ecotoxicological risk showed that the lower trophic level organisms were having adverse impact due to DDT and endosulfan residues in surface river water at certain locations of both Hooghly and Brahmaputra. Detected endosulfan showed

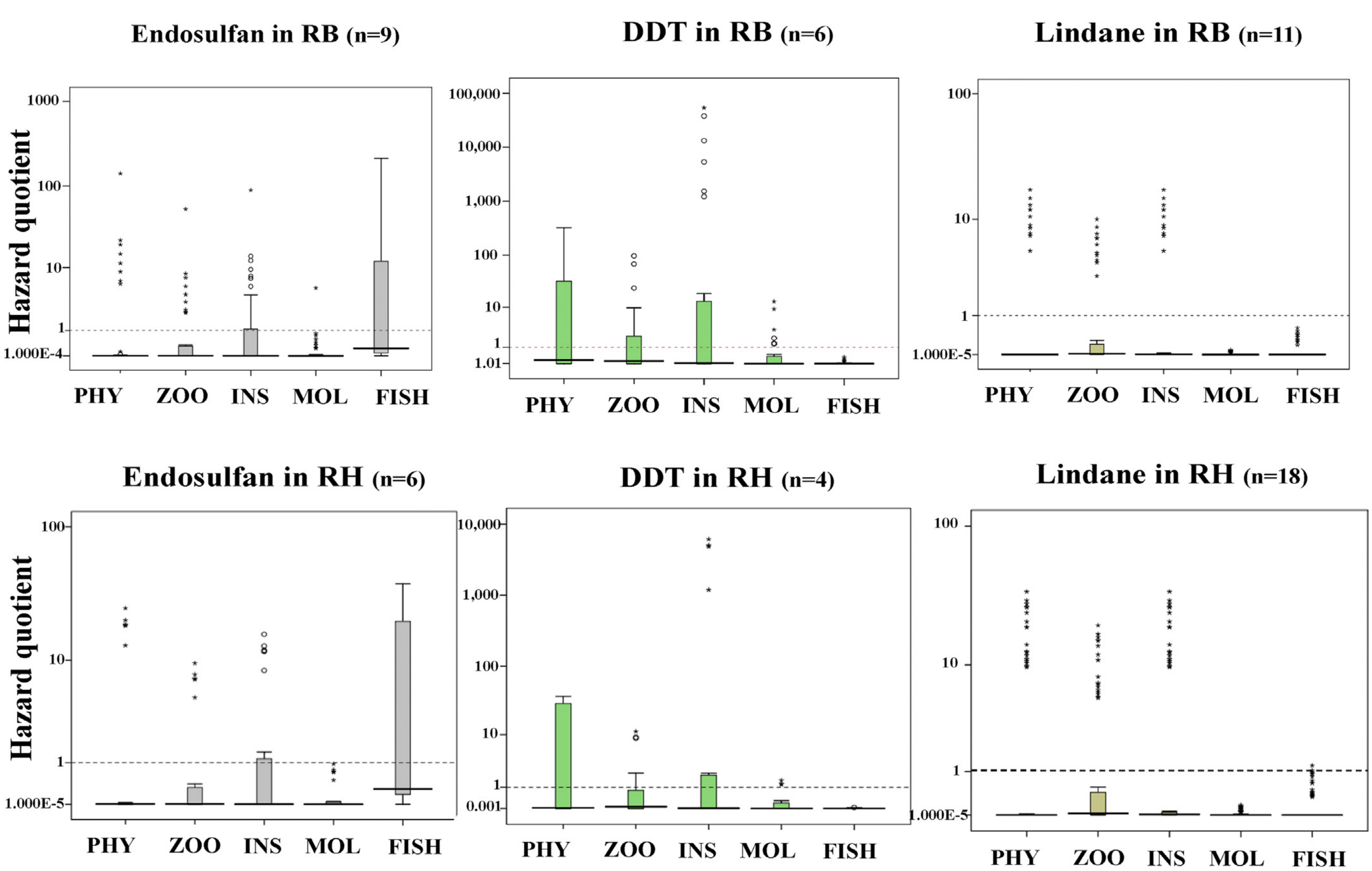

Fig. 4. Boxplot showing the range of ecotoxicological risk for organisms in different trophic levels viz., phytoplankton (PHY), zooplankton (ZOO), insect (INS), mollusc (MOL) and fish due to detected concentrations of endosulfan, DDT and lindane in River Brahmaputra (RB) and River Hooghly (RH).

Please cite this article in press as: Chakraborty, P., et al., Polychlorinated biphenyls and organochlorine pesticides in River Brahmaputra from the outer Himalayan Range and River Hooghly emptying into the Bay of Bengal: Occurrence, sources and ecotoxicological risk assessment, Environmental Pollution (2016), http://dx.doi.org/10.1016/j.envpol.2016.06.067

adverse impact on fish species hence the possibility of its impact on food chain cannot be ignored.

Acknowledgement

This work was supported by the support of SRM University start-up research grant and the Fast Track Research Grant under Young Scientist Scheme funded by the Department of Science and Technology, Government of India (SR/FTP/EE-44/2012). Authors would like to thank Mr. Elvis Dsouza managing partner of EDPC polymer industries Mumbai, for his extensive support during sampling campaign.

Appendix A. Supplementary data

Supplementary data related to this article can be found at http://dx.doi.org/10.1016/j.envpol.2016.06.067

References

Agriculture, D.O., 2013. Agriculture at a Glance. Department of Agriculture and Co-operation (Horticulture division), New Delhi.
AMAP, 2014. Trends in Stockholm Convention Persistent Organic Pollutants (POPs) in Arctic Air, Human Media and Biota, AMAP Technical Report to the Stockholm Convention. AMAP Technical ReportNo. 7. Arctic Monitoring and Assessment Programme (AMAP), Oslo, Norway.
Bhattacharya, B., Sarkar, S.K., Mukherjee, N., 2003. Organochlorine pesticide residues in sediments of a tropical mangrove estuary, India: implications for monitoring. Environ. Int. 29, 587–592.
Chakraborty, P., Sakthivel, S., Kumar, B., Kumar, S., Mishra, M., Verma, V., Gaur, R., 2014. Spatial distribution of persistent organic pollutants in the surface water of River Brahmaputra and River Ganga in India. Rev. Environ. Health 29, 45–48.
Chakraborty, P., Zhang, G., Eckhardt, S., Li, J., Breivik, K., Lam, P.K., Tanabe, S., Jones, K.C., 2013. Atmospheric polychlorinated biphenyls in Indian cities: levels, emission sources and toxicity equivalents. Environ. Pollut. 182, 283–290.
Chakraborty, P., Zhang, G., Li, J., Selvaraj, S., Breivik, K., Jones, K.C., 2016. Soil concentrations, occurrence, sources and estimation of air–soil exchange of polychlorinated biphenyls in Indian cities. Sci. Total Environ. http://dx.doi.org/10.1016/j.scitotenv.2016.03.009.
Chakraborty, P., Zhang, G., Li, J., Sivakumar, A., Jones, K.C., 2015. Occurrence and sources of selected organochlorine pesticides in the soil of seven major Indian cities: assessment of air–soil exchange. Environ. Pollut. 204, 74–80.
Chakraborty, P., Zhang, G., Li, J., Xu, Y., Liu, X., Tanabe, S., Jones, K.C., 2010. Selected organochlorine pesticides in the atmosphere of major Indian cities: levels, regional versus local variations, and sources. Environ. Sci. Technol. 44, 8038–8043.
Chi, K.H., Chang, M.B., Kao, S.J., 2007. Historical trends of PCDD/Fs and dioxin-like PCBs in sediments buried in a reservoir in Northern Taiwan. Chemosphere 68, 1733–1740.
ChinaSEPA, 2002. Environmental Quality Standards for Surface Water, Beijing.
CPCB, 2008. Performance of Sewage Treatment Plants – Coliform Reduction. Control of Urban Pollution. Central Pollution Control Board, Ministry of Environment & Forests. http: www.cpcb.nic.in.
Devi, N., Raha, P., 2013. Contamination of organochlorine pesticides (OCPs) in India. Bull. Environ. Sci. Res. 2, 9–14.
Devi, N.L., Qi, S., Chakraborty, P., Zhang, G., Yadav, I.C., 2011. Passive air sampling of organochlorine pesticides in a northeastern state of India, Manipur. J. Environ. Sci. 23, 808–815.
Devi, N.L., Yadav, I.C., Shihua, Q., Chakraborty, P., Dan, Y., 2014. Distribution and risk assessment of polychlorinated biphenyls (PCBs) in the remote air and soil of Manipur, India. Environ. Earth Sci. 72, 3955–3967.
Eqani, S.A.-M.-A.-S., Malik, R.N., Katsoyiannis, A., Zhang, G., Chakraborty, P., Mohammad, A., Jones, K.C., 2012. Distribution and risk assessment of organochlorine contaminants in surface water from River Chenab. Pak. J. Environ. Monit. 14, 1645–1654.
Erdogrul, Ö., Covaci, A., Schepens, P., 2005. Levels of organochlorine pesticides, polychlorinated biphenyls and polybrominated diphenyl ethers in fish species from Kahramanmaras. Turk. Environ. Int. 31, 703–711.
Fan, S., 2007. Endosulfan Risk Characterization Document. Environmental Monitoring Branch. Department of Pesticide Regulation, California Environmental Protection Agency, p. 51.
Fiedler, H., 2008. Stockholm convention on POPs: Obligations and Implementation, the Fate of Persistent Organic Pollutants in the Environment. Springer, pp. 3–12.
Gao, J., Liu, L., Liu, X., Lu, J., Zhou, H., Huang, S., Wang, Z., Spear, P.A., 2008. Occurrence and distribution of organochlorine pesticides–lindane, p, p′-DDT, and heptachlor epoxide–in surface water of China. Environ. Int. 34, 1097–1103.
GEFIO, 2002. Regionally Based Assessment of Persistent Toxic Substances – Indian Ocean Regional Report. Global Environment Facility, United Nations Environmental Programme, Geneva. http://www.chem.unep.ch/Pts/.
Guan, Y.-F., Wang, J.-Z., Ni, H.-G., Zeng, E.Y., 2009. Organochlorine pesticides and polychlorinated biphenyls in riverine runoff of the Pearl River Delta, China: assessment of mass loading, input source and environmental fate. Environ. Pollut. 157, 618–624.
Guide, G.C., 2011. The Story of Endosulfan available at: http://greencleanguide.com/the-story-of-endosulfan-in-india/.
Gupta, P., 2004. Pesticide exposure—Indian scene. Toxicol 198, 83–90.
Gurusubramanian, G., Rahman, A., Sarmah, M., Roy, S., Bora, S., 2008. Pesticide usage pattern in tea ecosystem, their retrospects and alternative measures. J. Environ. Biol. 29, 813–826.
Guzzella, L., Roscioli, C., Vigano, L., Saha, M., Sarkar, S., Bhattacharya, A., 2005. Evaluation of the concentration of HCH, DDT, HCB, PCB and PAH in the sediments along the lower stretch of Hugli estuary, West Bengal, northeast India. Environ. Int. 31, 523–534.
ICAR, 2016. State Specific- West Bengal. Indian Council of Agricultural Research available at: http://www.icar.org.in/files/state-specific/.
Jorgenson, J.L., 2001. Aldrin and dieldrin: a review of research on their production, environmental deposition and fate, bioaccumulation, toxicology, and epidemiology in the United States. Environ. Health Perspect. 109, 113.
Kang, J.-H., Choi, S.-D., Park, H., Baek, S.-Y., Hong, S., Chang, Y.-S., 2009. Atmospheric deposition of persistent organic pollutants to the east Rongbuk glacier in the himalayas. Sci. Total Environ. 408, 57–63.
Kaushik, A., Sharma, H., Jain, S., Dawra, J., Kaushik, C., 2010. Pesticide pollution of river Ghaggar in Haryana, India. Environ. Monit. Assess. 160, 61–69.
Kaushik, C., Sharma, H., Jain, S., Dawra, J., Kaushik, A., 2008. Pesticide residues in river Yamuna and its canals in Haryana and Delhi, India. Environ. Monit. Assess. 144, 329–340.
Kumar, B., Kumar, S., Sharma, C.S., 2013. Ecotoxicological risk assessment of polychlorinated biphenyls (PCBs) in bank sediments from along the Yamuna River in Delhi, India. Hum. Ecol. Risk Assess. Int. J. 19, 1477–1487.
Kumar, B., Singh, S.K., Mishra, M., Kumar, S., Sharma, C.S., 2012. Assessment of polychlorinated biphenyls and organochlorine pesticides in water samples from the Yamuna River. J. Xenobiotics 2, 6.
Kumar, S.C.A.A., 2010. Fishing Toxics-Mercury Contamination of Fish in West Bengal. Toxic links and DISHA, New Delhi, p. 125.
Kuranchie-Mensah, H., Atiemo, S.M., Palm, L.M.N.-D., Blankson-Arthur, S., Tutu, A.O., Fosu, P., 2012. Determination of organochlorine pesticide residue in sediment and water from the Densu river basin, Ghana. Chemosphere 86, 286–292.
Liu, G., Zheng, M., Cai, M., Nie, Z., Zhang, B., Liu, W., Du, B., Dong, S., Hu, J., Xiao, K., 2013. Atmospheric emission of polychlorinated biphenyls from multiple industrial thermal processes. Chemosphere 90, 2453–2460.
Malik, A., Ojha, P., Singh, K.P., 2009. Levels and distribution of persistent organochlorine pesticide residues in water and sediments of Gomti River (India)—a tributary of the Ganges River. Environ. Monit. Assess. 148, 421–435.
Mishra, K., Sharma, R.C., Kumar, S., 2012. Contamination levels and spatial distribution of organochlorine pesticides in soils from India. Ecotoxicol. Environ. Safe 76, 215–225.
Na, T., Fang, Z., Zhanqi, G., Ming, Z., Cheng, S., 2006. The status of pesticide residues in the drinking water sources in Meiliangwan Bay, Taihu Lake of China. Environ. Monit. Assess. 123, 351–370.
Nøst, T.H., Halse, A.K., Randall, S., Borgen, A.R., Schlabach, M., Paul, A., Rahman, A., Breivik, K., 2015. High concentrations of organic contaminants in air from ship breaking activities in Chittagong, Bangladesh. Environ. Sci. Technol. 49, 11372–11380.
NVBDCP, 2010. National Vector Borne Disease Control Programme, National Rural Health Mission. National Rural Health Mission, Bhopal.
PPIN, 2013. Statistics of Pesticides available at: http://ppqs.gov.in/IpmPesticides_Cont.htm.
Rajendran, R.B., Imagawa, T., Tao, H., Ramesh, R., 2005. Distribution of PCBs, HCHs and DDTs, and their ecotoxicological implications in Bay of Bengal, India. Environ. Int. 31, 503–512.
Ramaswamy, B.R., Shanmugam, G., Velu, G., Rengarajan, B., Larsson, D.J., 2011. GC–MS analysis and ecotoxicological risk assessment of triclosan, carbamazepine and parabens in Indian rivers. J. Hazard. Mater. 186, 1586–1593.
Rissato, S.R., Galhiane, M.S., Ximenes, V.F., De Andrade, R.M., Talamoni, J.L., Libânio, M., De Almeida, M.V., Apon, B.M., Cavalari, A.A., 2006. Organochlorine pesticides and polychlorinated biphenyls in soil and water samples in the Northeastern part of São Paulo State, Brazil. Chemosphere 65, 1949–1958.
Rudra, K., 2007. The Status of Water Resources in West Bengal (A Brief Report) available at: http://www.indiawaterportal.org/articles/status-water-resources-west-bengal-report-kalyan-rudra.
Sandrp, 2013. Brahmaputra – the Beautiful River or the Battleground? available at: https://sandrp.wordpress.com/2013/07/17/brahmaputra-the-beautiful-river-or-the-battleground/.
Selvaraj, K.K., Shanmugam, G., Sampath, S., Larsson, D.J., Ramaswamy, B.R., 2014. GC–MS determination of bisphenol A and alkylphenol ethoxylates in river water from India and their ecotoxicological risk assessment. Ecotoxicol. Environ. Safe 99, 13–20.
Semeena, V.S., Feichter, J., Lammel, G., 2006. Impact of the regional climate and substance properties on the fate and atmospheric long-range transport of persistent organic pollutants-examples of DDT and γ-HCH. Atmos. Chem. Phys. 6, 1231–1248.
Sharma, A., 2013. Stockholm Convention on persistent organic pollutants: challenges towards its implementation in India. Int. J. Res. Environ. Sci. Technol. 3, 117–121.

Sheng, J., Wang, X., Gong, P., Joswiak, D.R., Tian, L., Yao, T., Jones, K.C., 2013. Monsoon-driven transport of organochlorine pesticides and polychlorinated biphenyls to the Tibetan Plateau: three year atmospheric monitoring study. Environ. Sci. Technol. 47, 3199–3208.

Sibali, L.L., Okwonkwo, J.O., McCrindle, R.I., 2008. Determination of selected organochlorine pesticide (OCP) compounds from the Jukskei River catchment area in Gauteng, South Africa. Water SA 34, 611–622.

Singh, K.P., Malik, A., Sinha, S., 2007. Persistent organochlorine pesticide residues in soil and surface water of northern Indo-Gangetic alluvial plains. Environ. Monit. Assess. 125, 147–155.

Sun, C., Dong, Y., Xu, S., Yao, S., Dai, J., Han, S., Wang, L., 2002. Trace analysis of dissolved polychlorinated organic compounds in the water of the Yangtse River (Nanjing, China). Environ. Pollut. 117, 9–14.

Turgut, C., 2003. The contamination with organochlorine pesticides and heavy metals in surface water in Küçük Menderes River in Turkey, 2000–2002. Environ. Int. 29, 29–32.

USEPA, 1998. Guidelines for Ecological Risk Assessment. Available at: http://www.epa.gov/raf/publications/pdfs/ECOTXTBX.PDF.

USEPA, 2006. United States Environmental Protection Agency. National Recommended Water Quality Criteria (NRWQC) (4304T).

Weber, J., Halsall, C.J., Muir, D., Teixeira, C., Small, J., Solomon, K., Hermanson, M., Hung, H., Bidleman, T., 2010. Endosulfan, a global pesticide: a review of its fate in the environment and occurrence in the Arctic. Sci. Total Environ. 408, 2966–2984.

Webster, P., 2004. Study finds heavy contamination across vast. Russ. Arct. Sci. 306, 1875–1875.

Zhou, R., Zhu, L., Chen, Y., 2008. Levels and source of organochlorine pesticides in surface waters of Qiantang River, China. Environ. Monit. Assess. 136, 277–287.

Further reading

Yu, Y., Li, Y., Shen, Z., Yang, Z., Mo, L., Kong, Y., Lou, I., 2014. Occurrence and possible sources of organochlorine pesticides (OCPs) and polychlorinated biphenyls (PCBs) along the Chao River, China. Chemosphere 114, 136–143.