Environmental Pollution 263 (2020) 114229

Contents lists available at ScienceDirect

Environmental Pollution

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

First account of spatio-temporal analysis, historical trends, source apportionment and ecological risk assessment of banned organochlorine pesticides along the Ganga River☆

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A R T I C L E I N F O

Article history:
Received 23 July 2019
Received in revised form
15 February 2020
Accepted 16 February 2020
Available online 4 March 2020

Keywords:
Ganga river
Organochlorine pesticides
Spatial distribution
Seasonal variation
Source apportionment
Ecological risk assessment

A B S T R A C T

We conducted the first comprehensive assessment of the presence, source, and ecotoxicological implication of 13 banned and restricted organochlorine pesticides (OCPs) in the surface water along the Ganga River for two different seasons. Surface water samples were collected along the 2525 km stretch of the Ganga through 43 sites representing five zones of diverse land-use pattern, pesticide consumption rate, and varied flow. The mean concentrations of ΣOCPs were significantly higher (~2–5 times) in the post-monsoon or wet season [range: 0.126 to 10.402 μg/L (mean: 2.482 μg/L ± 3.589 and median: 1.433)] than in the post-winter or dry season [range: 0.053 to 3.010 μg/L (mean: 0.765 μg/L±1.033 and median: 0.399)]. Lindane (γ-HCH) was the dominant and most frequently detected pesticide at all the sites, indicating possible continued use of this banned pesticide in agricultural practices. The spatial distribution of OCPs revealed non-significant difference amongst different zones and indicate that point source pollution from the open drains along the Ganga could be responsible for observed trend. Ratio diagnostic analysis highlighted the fresh inputs and potential illegal use of lindane and chlordane at all the zones whereas, historical use of DDT was revealed at the majority of sites. Interestingly, fresh inputs of DDT were observed in the relatively pristine high altitude Upper zone (UZ) suggesting long-range atmospheric transfer and its continued use in the zone. Risk quotient (RQ) analysis revealed high ecotoxicological risks (>1), at all the studied sites for p, p' DDE. The lower zone (LZ) emerged as a high ecological risk zone. The study highlights that though the OCPs analysed in this study are banned/restricted in India, still the implementation of the ban is poor and delayed and the country requires stricter adherence to its National Implementation Plan (NIP) on pesticides.

1. Introduction

Agriculture, along with its associated sectors, is the primary source of livelihood for 58% of the population in India and plays a central role in ensuring the steady growth of the Indian economy. One of the pivotal contributors toward food security and accelerated agricultural yields is the use and application of pesticides and other chemicals. Consequently, the agrochemical industry in India has witnessed an outstanding growth. However, high pesticide applications have also led to the accumulation of these chemicals in the environment through several routes (Schulz, 2001; Gavrilescu, 2005; Holvoet et al., 2007; Tanaka and Katagi, 2008), which has the potential to affect ecosystem and its services" (Tanaka and Katagi, 2008; Carriquiriborde et al., 2014).

Among pesticides, organochlorines pesticides (OCPs) cause serious concern owing to their persistence, toxicity, long-range transmission, and bio-accumulative nature (Naqvi and Vaishnavi, 1993; Willett et al., 1998; Contreras López, 2003 Briz et al., 2011; Gao et al., 2013). In human beings, studies have indicated possible relationships between OCPs and various cancers (Mathur et al., 2002; Rathore et al., 2002; Abdo et al., 2013; Louis, 2019; Ennour-Idrissi et al., 2019), teratogenicity (Kalra et al., 2016; Kim et al., 2017; Ramakrishnan and Jayaraman, 2019), endocrine disruption (Younglai et al., 2004; Fowler et al., 2007; Frye et al., 2012), neurotoxicity (Shinomiya and Shinomiya, 2003; Sharma et al., 2010; Song et al., 2012; Heusinkveld and Westerink, 2012), and genotoxicity (Pandey et al., 1990; Ramírez and Cuenca, 2002; Poli

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☆ This paper has been recommended for acceptance by Christian Sonne.

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https://doi.org/10.1016/j.envpol.2020.114229
0269-7491/

et al., 2003; Ennaceur et al., 2008). Similarly, wildlife species exposed to OCPs have shown high rates of malformed genitalia (Sonne et al., 2006), aberrant mating behavior (Fry, 1995), sterility, cancer, egg-shell thinning, and immune system and thyroid dysfunction (Colborn and Smolen, 1996; Briz et al., 2011; Bergman et al., 2012; Newton, 2013; WHO, 2013; Godfray et al., 2019).

Until the year 2006, when India ratified the Stockholm Convention, large amounts of OCPs banned by other countries were still manufactured, used, and exported. Post-ratification, India banned the manufacturing, import, export, and use of all of the 12 persistent organic pollutants (POPs) listed in the convention, except for dichlorodiphenyltrichloroethane (DDT). India is the only major country worldwide to still manufacture and use DDT for vector control and is allowed exemption until 2022. Specific exemption (up to the year 2019) was also allowed for its use as an intermediate in the production of Dicofol (another OCP used against mites). The country also exports DDT to several countries either as a pesticide or as a chemical intermediate (Toxics Link, 2018). However, the production of DDT has declined in the last decade, from 3310 MT in 2008–2009 to 1370 MT in 2018–2019 (Annual Reports, Dept. of Chemicals and Petrochemicals, 2011–2019). Among hexachlorocyclohexanes (HCHs), only lindane (γ-HCH) is banned for production, import, formulation (since March 2011), and use (since March 2013). However, India does not have any restriction on α-, β-, and δ-HCHs (PPQS, 2019). The manufacture, sale, use, and export of endosulfan has been banned since May 2011; however, data suggest exportation of endosulfan from India still occurs. (Dept. of Commerce, India, 2019). The supreme court of India had allowed export of stockpiles of endosulfan, which is lying in stock unsold as an inventory, to other countries that were interested (Interim report, Joint expert Committee, 2011). A summary of other OCPs listed as POPs under the Stockholm Convention and thus banned in India is given in Table S1.

Despite the imposed ban, over the last two decades, high levels and distributions of OCPs have frequently been detected across many rivers in India (Table 3 & Table S10), including the large rivers (Table: S11). Hence, the investigation of OCPs in aquatic environments especially large rivers is the primary source of information to assess the actual status of the pesticide ban policy in the country. The concentrations serve as an indicator of contaminant load and anthropogenic impact on the environment.

For our study, we chose the Ganges River (Ganga), the second-largest river in India supporting 36.1% of the country's population (438.2 million), about 70% of which is dependent on agriculture (Census, 2011). The Ganga has exceptional value in the country owing to its religious, economic, and ecological significance. During its course, from origin to outfall, the Ganga is joined by several tributaries to form the vast and highly fertile Indo-Gangetic plains, which has given rise to intensive agriculture practices along the river (Karnick, 2011). The cumulative consumption of pesticides in Ganga states between 2012 and 2017 was 72,741 metric tons, accounting for 27% of the total pesticide consumption in the country (PPQS, 2017).

Previous studies on the Ganga have shown OCPs to be the dominant group of pesticides detected in selected stretches of the river (Rehana et al., 1995; Nayak et al., 1995; Sankararamakrishnan et al., 2005; Ghose et al., 2009; Singh et al., 2012; Mutiyar and Mittal, 2013; Raghuvanshi et al., 2014; Chakraborty et al., 2016; and Mondal et al., 2018). These studies have mostly focused on DDT, HCH, and endosulfan occurrence in selected stretches without considering the status of other banned pesticides or the full spatial coverage of the river. Besides, assessment of the ecological risks to aquatic species, temporal trends, and possible sources, are also lacking in these studies. The lack of a comprehensive study has affected biodiversity conservation plans and environmental monitoring efforts for the Ganga. Consequently, further research is needed to improve the current information on OCP pollution in the Ganga and create baseline data on the status of banned OCPs across all five Ganga states.

In this paper, we aim to investigate the occurrence, spatiotemporal distribution, potential sources, and ecological risks of persistent and banned OCPs to aquatic species along all the five Ganga states. To the best of our knowledge, this is the first comprehensive study on Ganga to include the above four criteria. This study is a part of the project “Biodiversity Conservation and Ganga Rejuvenation” aimed at developing comprehensive measures to conserve and safeguard the aquatic biodiversity of the Ganga. The results from this study will be helpful to the policy-makers in identifying the status and ecological risks of banned pesticide usage at the state level. This in turn will help in designing effective monitoring strategies and formulating appropriate pesticide mitigation efforts that would be directed toward sustaining the healthy ecological diversity of the Ganga.

2. Material and methods

2.1. Study area

The Ganga lies between north latitudes 22°1′43.284″–30°49′59.99″ and east longitudes 79°09′60.00″–88°12′37.584″. The drainage in the Ganga is a combination of precipitation, tributaries, and snowmelt water from the Himalayas. It covers a distance of 2525 km as it flows through five states from the north to the east of the country (Fig. 1).

The catchment of the Ganga is an area of intensive agriculture activity. The Ganga states has a high net irrigation to sown ratio (62.6%) compared to that of the entire country (44%) ESMF (2011). The major crops cultivated in the five Ganga states are either Rabi or Kharif. The Kharif crops such as rice, maize, and sugarcane are sown in June and July and are harvested in September and October, whereas the Rabi crops such as wheat, pulses, and peas are sown in the period between October and December and harvested in April and May. During the dry season (January to May), the riverbed cultivation of seasonal vegetables is a common practice in the dry riverbeds of the Ganga (Uttar Pradesh State Biodiversity Board, 2013; Kumari et al., 2018). The farmers of the Ganga belt practice multiple cropping pattern; hence, the fields are irrigated more than once a year.

Ganga is also home to a rich and diverse fauna, including endangered species, such as the dolphin *Platanista gangetica*; the otters *Lutrogale perspicillata, Lutra lutra*, and *Aonyx cinereus*; the gharial *Gavialis gangeticus*; the crocodiles *Crocodylus palustris* and *Crocodylus porosus*; the turtle *Batagur kachuga*; and the fishes *Tor putitora* and *Tenualosa ilisha* (WII, 2018).

Along its course, the Ganga receives 139 drains, discharging 6087 MLD (million litre per day) of municipal and industrial waste daily and it is reported that there is a gap of 80% between generated wastewater and treatment capacity (CPCB, 2013). Both point source (drains) and non-point source (agriculture) of pollution could severely affect the water quality, as well as the life and health of the species—including humans—depending on it.

2.2. Sampling sites

Among the five Ganga states, there is a significant difference in the environmental flows, demographics, average pesticide consumption rates, and land-use patterns (Table 1 & Table S8). Therefore, based on the key features presented in Table 1, we demarcated the 2525 km stretch of the Ganga into five homogenous zones (Fig. 1 & Table 1), to gain a better understanding of the

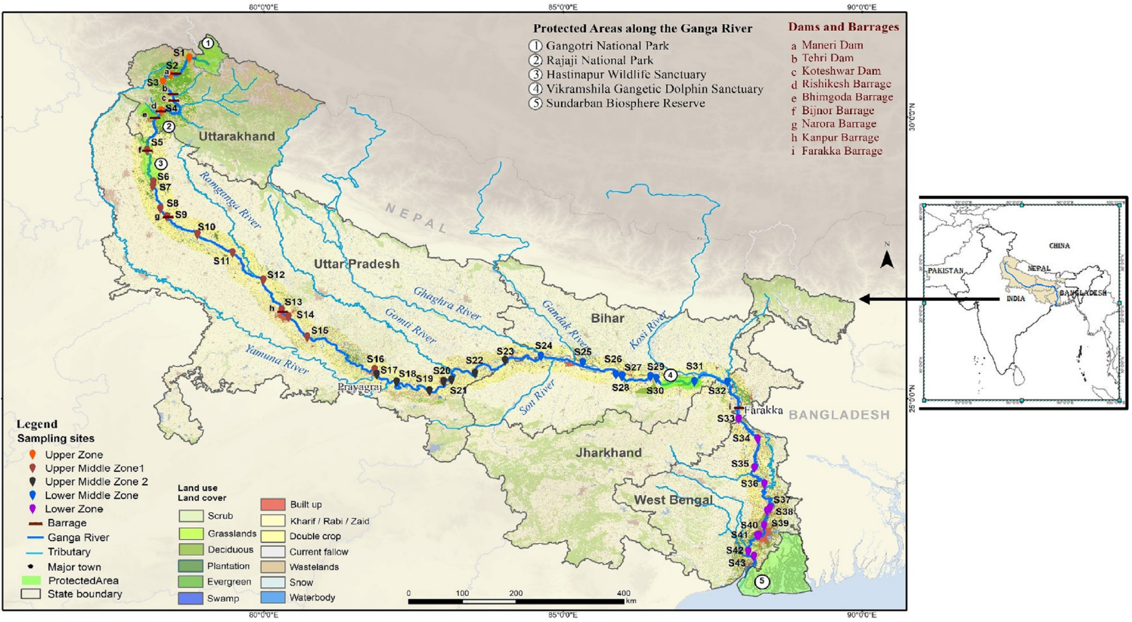

Fig. 1. Representation of sampling sites along Ganga River and segregation into different zones.

Table 1
Demarcation of zones based on demographics, cultivable land, pesticide consumption, and local precipitation in the states of the Ganga.

States	River length (Km)	Geographical Area (Sq Kms)	Demography (Billions) Census (2011) [a]	Total cropped Area (000 Thousand Hectares, 2013–2014)[b]	Cropping Intensity# (%)\ (2013–2014)[b]	Total pesticide consumption (%) 2016–2017[c]	Annual precipitation		Zone Demarcation
							2016	2017[d]	
Uttarakhand	450	53483	0.01	1099	146	0.92	1308.6	1476	UZ
Uttar Pradesh	1000	240928	0.2	25896	145	71.03	692.8*	585.2*	UMZ1+ UMZ2++
Bihar	400	94163	0.1	7580	123	5.88	1158	1112	LMZ
Jharkhand	45	79716	0.03	1672	59	3.79	1264	1165.8	
West Bengal**	540	88752	0.09	9618	172	18.38	1427	1568.6	LZ

*Average of total annual rainfall in East Uttar Pradesh and West Uttar Pradesh. **Ganga River in this state/zone is also known as Hooghly River.
+UMZ1 = Before Confluence with Yamuna River.
++UMZ1 = After Confluence with Yamuna River.
#Cropping Intensity = Gross Cropped Area x 100/Net Sown Area.
[a] Census of India Office.
[b] Directorate of Economics and Statistics.
[c] PPQS (2017).
[d] data.gov.in/catalog/rainfall-india.

factors influencing the distribution of OCPs in its different stretches. These five zones are identified as the Upper Zone (UZ), Upper Middle Zone 1 (UMZ1; before the confluence with the Yamuna River), Upper Middle Zone 2 (UMZ2; after the confluence), Lower Middle Zone (LMZ), and Lower Zone (LZ).

Within these five zones, 43 sampling sites (Table S2) were selected for detailed characterization of the OCP pollution dynamics. These sites were selected based on the presence/absence of agricultural and industrial belts, identifying gaps through detailed literature references from previous studies and considering the existing monitoring stations prescribed by the Central Pollution Control Board of India. Taking the above criteria into account, we conducted a rapid reconnaissance survey between May and June, 2017, to finalize the sampling sites. Subsequently, samples were collected throughout two seasons (post-monsoon or wet season [October to November 2017] and post-winter or dry season [April to May 2018]) to characterize the influence of seasonal variation on the water quality.

2.3. *Pesticide sampling method and analysis*

At each sampling site (n = 43), three surface water samples were collected randomly, at an average depth of 30 cm, in amber-colored pre-cleaned high-density polyethylene (HDPE) bottles, which were bulked together to form a composite sample. After collection, samples were stored in an ice box for shipping to the laboratory and kept at 4 °C until analysis (USGS, 2006; CPCB, 2007).

2.3.1. *Target pesticides*

Thirteen OCPs investigated in this study are selected on the basis of their high historic usage, detection frequency reported in previous studies (Table 3), pesticides included in the CPCB- India monitoring network (CPCB, 2013), OCPs listed as POP under Stockholm convention (UNEP (2012)), and, OCPs with endocrine disrupting properties (Ratcliffe, 1970; Schreiber and Risebrough, 1972; Gress et al., 1973; Helle, 1976; Fry and Toone, 1981; Cranmer et al., 1984; Cummings and Gray, 1987; Roy Chowdhury et al., 1987; Gray et al., 1989; Guillette et al., 1994; Willingham and Crews, 1999; Guillette et al., 2000; Huang et al., 2004a,b; Bergman et al., 2012). The target analytes are:

p,p′-dichlorodiphenyltrichloroethane(p,p′-DDT), p,p′-dichlorodiphenyldichloroethane (p,p′-DDD), p,p′-dichlorodiphenyldichloroethylene (p,p′-DDE), alpha-hexachlorocyclohexane (α-HCH), beta-hexachlorocyclohexane (β-HCH), gamma-hexachlorocyclohexane (γ-HCH) or lindane, delta hexachlorocyclohexane (δ-HCH), Chlordanes (CHLs) namely cis-chlordane (c-CHL), and trans-chlordane (t-CHL), Methoxychlor (M-CHLR), Endosulfan (ES) namely α-endosulfan (α-ES), and β-endosulfan (β-ES) and, endosulfan sulfate (ESS).

2.3.2. Chemicals and instruments

The OCP standard mixture solution containing 13 target analytes with >99% purity was purchased from Sigma-Aldrich (USA). A standard OCP stock solution (500 mg/l) was prepared with hexane and stored in dark at 2 °C before use. All the solvents and chemicals used in the extraction process were chromatographically pure grade (Merck, USA). A Shimadzu Model 2010 Gas Chromatograph (GC) equipped with 63Ni Electron Capture detector (ECD) was used for detection and quantification of OCP residues.

2.3.3. OCP extraction and analysis

Following flow diagram shows the extraction method, performed as recommended by EPA method 1699 (EPA, 2007) with slight modifications:

1 L Water → Filtration (0.45 μm filter)→ Liquid-Liquid Extraction ((twice with DCM − 35 ml) & (final Hexane −25 ml & 7.5 ml NaCl))→ Anhydrous Na2SO4 (5 gm) → Drying → Reconstitution in 1 ml n-Hexane → GC-ECD (Rtx-5 (30 m × 0.25 mm × 0.25 μm, Restek)

The GC programme was set as follows:

Injector and detector temperature - 200 °C and 320 °C respectively; Oven temperature (Initial) 100 °C (1 min) → 180 °C at 25 °C/min (2 min), ramp 5 °C/min → 240 °C (1 min), and at 4.5 °C/min to 260 °C (2 min) → at 10 °C/min to 280 °C (5 min).

Furthermore, 20% of samples were validated and confirmed by GC-MS/MS results with RSD ≤ 10%. The samples were send to the Shimadzu Analytical Lab, India for GC MS/MS analysis.

2.4. Quality assurance and quality control (QA/QC)

QA/QC was performed according to the requirements of ISO/IEC 17025 and US EPA guidelines. Field, laboratory blank, and solvent blank samples were run routinely to check for interferences and cross-contamination. The blank concentrations were less than the method detection limit (<MDL). Linear calibration curves were obtained with the r^2 value of 0.999 and calibration verification (standard deviation) was less than 5%. The target compound concentrations were quantified by an external standard method using the peak height and area of the standards at five level calibration curves. A matrix spike recovery study was performed by spiking the samples with known working standard solutions of OCPs and extracted and analysed in the same way as the real samples. The matrix spike recoveries were in the range of 82%–124% for the studied compounds. After every 10 samples, a standard quality check was performed. The results of the analysis are reported in μg/L and Schimadzu Lab Solution Software was used for data acquisition (Tables S3 and S4). MDLs were assessed based on US Environmental Protection Agency (EPA) guidelines (USEPA, 2016) and the minimum levels of quantification were determined based on concentrations 10 times the detected MDL. Any target analyte detected below the MDL was considered as below detection limits (<DL).

2.5. Potential ecological risk assessment

An ecological risk assessment across the stretch of the Ganga was conducted using the ecological risk quotient (RQ) model. RQ is established based on Eq. (1).

$$RQ = \frac{MEC}{PNEC} \tag{1}$$

where, MEC is the mean or maximum measured environmental concentration and PNEC is the predicted no-effect concentration. PNEC is derived from the lowest toxicity value (i.e., no-observed effect concentration (NOEC) value) observed for the most sensitive species. When NOEC values were not available, we used LC50 or EC50 values after correction by an assessment factor intended to extrapolate from acute to chronic toxicity and for removing the uncertainty arising from the extrapolation from intra- and inter-species variability in sensitivity (Table S6). (European Commission, 2000) based on Eq. (2).

In this assessment, the respective NOEC, LC50 or EC 50 values for three trophic levels (primary producers i.e., algae; primary consumers i.e., aquatic invertebrates; secondary consumers i.e., fish) were used to determine the PNEC.

$$PNEC = \frac{NOEC\ or\ LC50\ orEC50}{Assessment\ Factor} \tag{2}$$

For most pesticides, NOEC/LC50/EC50 values were obtained from the Pesticides Properties DataBase (PPDB) whereas others were taken from US EPA ECOTOXicology knowledgebase (ECOTOX). Furthermore, to identify the high-risk zones, the average and maximum detected concentrations at each zone were used for determination of the general (RQm) and worst-case (RQex) scenarios, respectively.

The risk to aquatic species, based on risk ratios, was subsequently classified into four risk levels comprising high, medium, low, and negligible ecological risks, corresponding to RQ values ≥ 1, 0.1–1, 0.01–0.1, and <0.01, respectively (Sánchez-Bayo et al., 2002; Palma et al., 2014; Zhang et al., 2016).

2.6. Statistical analysis and Softwares

The sampling data were tested for normality using the Kolmogorov-Smirnov test. As the distribution was non-normal therefore, Kruskal-Wallis (KW) with Dunn's post-hoc was applied to test significant differences among zones whereas Mann-Whitney *U* test (MWU) was conducted to test the temporal differences. Differences were considered significant at $p < 0.05$. Statistical analyses were performed using the statistical software SPSS (window version 23.0.).

Spatial distribution of OCPs along the Ganga River was analysed with the help of ArcGIS 10.6 (Esri).

3. Results and discussions

3.1. Occurrence and seasonal distribution of OCPs

A summary of the descriptive analysis of the 13 target OCPs detected in the Ganga is presented in Table 1. The total OCP concentrations (ΣOCPs) in water samples ranged from 0.126 to 10.402 μg/L (mean: 2.482 μg/L; median: 1.433 μg/L) in the post-monsoon season, and 0.053–3.010 g/L (mean: 0.765 μg/L; median: 0.399 μg/L) in the post-winter season. The mean concentrations of ΣOCPs were significantly (MWU test, U = 621, p = 0.009) higher (~2–5 times) in the post-monsoon season than in the post-winter season (Fig. 2). This could be explained by the high atmospheric

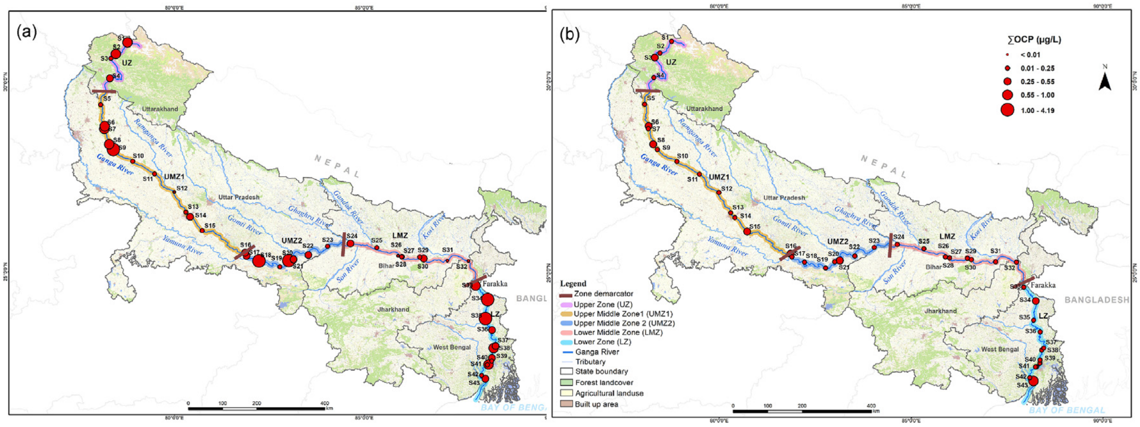

Fig. 2. Seasonal and spatial distribution of total OCPs in surface water samples of Ganga River in (a) Post-Monsoon season (b) Post-Winter season.

precipitation and subsequently higher agricultural surface runoff in the monsoon season, which facilitates the entry of pesticides into nearby waterbodies. This view is further confirmed by previous studies conducted in different river bodies of India (Kumarasamy et al., 2012; Singh et al., 2012; Alam et al., 2015; Maurya and Malik, 2016) and worldwide (Tanabe et al., 2001; Zhang et al., 2004; Yang et al., 2004; Li et al., 2010; Toan et al., 2013; Chen et al., 2018; Sun et al., 2018; Wang et al., 2018). In addition, just before the monsoon, approximately 5 km of land on both sides of the river bank in all the zones except for UZ is utilized extensively for the dry riverbed cultivation of seasonal vegetables, with possible high pesticide applications. These pesticides will likely enter surface water as runoff because of the high flooding and precipitation in the monsoon season, thus increasing their levels in the surface water of the river.

The most frequently detected OCPs (Detection Frequency (DF > 60%) in both the seasons were γ-HCH (DF 74%–95%) and α-HCH (DF 63%–95%). The lowest detection rate in both seasons was observed for isomers of endosulfan (α and β), with frequencies ranging between 9% and 20%.The groupwise mean concentration of OCPs in Ganga River for post-monsoon season followed the order ΣHCHs > ΣCHLs> ΣDDTs > ΣESs > M-CHLR> ΣESS. In the post-winter season, the OCP concentration followed the order ΣHCHs > ΣDDTs > ΣCHLs > ΣESs > M-CHLR > ΣESS. The lower detection rate and relatively low concentrations of M-CHLR and ESS could be attributed to their low usage in the past and relatively fast degradation in the water compartment (Table S1). The mean concentration of ΣHCHs in the Ganga River was approximately five times higher (*MWU test, U = 584, p = 0.003*) and that of CHL 1.4× higher (*MWU test, U = 907, p = 0.88*) in the post-monsoon season than in the post-winter season. The mean concentration of DDTs was found to be 1.5 × higher (*MWU test, U = 696.5, p = 0.003*) while ΣESs (*MWU test, U = 715.5, p = 0.03*), M-CHLR (*MWU test, U = 670.5, p = 0.08*), and ESS (*MWU test, U = 598, p = 0.001*) was 2.5-3x higher in the post-winter season than in the post-monsoon season. The high concentration levels of these OCPs during the post-winter season may be attributed to higher evaporation (low water level) and lower flow volume conditions in this season. Our findings are consistent with the results of previous studies conducted in various rivers of India (Malik et al., 2009; Raghuvanshi et al., 2014; Alam et al., 2015; Mondal et al., 2018).

3.1.1. Seasonal composition of OCPs

The compositional analysis of HCHs, DDTs, CHLs, and ESs for the post-monsoon and post-winter seasons are presented in Fig. S1. The compositional profile of HCHs revealed the dominance of the γ-HCH isomer with 71% in the post-monsoon and 43% in the post-winter season, followed by α-HCH (~23% for both seasons). The contribution (%) of two other HCH isomers, namely β-HCH and δ-HCH, toward ΣHCHs was found to be in the range of 1.03%–5% (post-monsoon) and 13%–17% (post-winter). Despite the ban, the widespread occurrence and elevated concentrations of γ-HCH residues could be attributed to their high historic usage (because of their low cost and broad spectrum insecticidal property), strong persistence, and potential fresh inputs (possibly through illegal sources) to the river. In addition, compared to other target OCPs that were banned years ago, γ-HCH was banned relatively recently (2013) in India (PPQS, 2019).

The percentage composition of DDTs revealed the dominance of *p,p′*-DDE (post-monsoon: 73%; post-winter: 48%), a breakdown product of *p,p′*-DDT, in the surface water of the Ganga that could be attributed to their slow degradation (Table S1) in water. *p′,p′*-DDD was found to be 11% in the post-monsoon season and 27% in the post-winter season whereas *p′,p′*-DDT contributed 16% in the post-monsoon and 25% in the post-winter season. The relative higher detection frequency (Table 2) of *p,p′*-DDD and *p,p′*-DDT in the post-winter season as compared to the post-monsoon season could be because of low dilution during the post-winter season resulting in their high concentration. Further, DDT is still being used in India during the post-winter season for malaria vector control. Therefore, higher concentration of *p,p′*-DDT could also be attributed to the fresh sprays of DDT.

Among the isomers of chlordane, t-CHL was dominant during both seasons (post-monsoon: 82%; post-winter: 54%), whereas among endosulfans, β-ES was identified as the dominant isomer (post-monsoon: 60%; post-winter: 84%).

3.2. Zone wise distribution of OCPs

Fig. 3 show the spatiotemporal dynamics and zone-wise comparison of ΣOCPs in the Ganga.

The zone-wise pollution gradient observed in the post-monsoon season was LZ > UMZ2 > UZ > UMZ1 > LMZ (Table S7).

The highest concentration of OCPs in the LZ (representing the state of West Bengal), in the post-monsoon season could be explained by the high cropping intensities in this catchment (Table 1), reduction in river flow (because of the diversion of river water to Bangladesh, and abstraction of water for irrigation) at Farakka Barrage (Table S8) and emergence of estuarine zone before the river enters the bay of bengal.

In the post-winter season, the OCPs followed the distribution pattern UMZ1 > LZ > UMZ2 > UZ > LMZ (Table S7). In the UMZ1, a substantial amount of water is diverted from the river to support agricultural activities through a system of canals, which may

Table 2
Concentration (μg/L) and detection frequency (%) rate of OCPs in water samples from the post-monsoon ($n = 43$) and the post-winter season ($n = 43$) for the Ganga.

Analyte	Post-Monsoon, October to November 2017 (n = 43)					Post-Winter, March to April 2017 (n = 43)				
	Range	Median	Mean	SD	DF%	Range	Median	Mean	SD	DF%
α-HCH	0.006–3.860	0.091	0.432	1.172	63	0.017–0.255	0.087	0.108	0.088	95
β- HCH	0.102–0.847	0.465	0.024	0.036	33	0.015–0.231	0.082	0.098	0.080	91
γ- HCH	0.004–4.346	0.812	1.722	1.888	74	0.016–1.137	0.111	0.233	0.367	95
δ - HCH	<DL -0.252	0.004	0.088	0.110	21	0.001–0.253	0.027	0.058	0.089	58
ΣHCH	**0.112–9.304**	**1.371**	**2.266**	**3.206**		**0.050–1.877**	**0.307**	**0.497**	**0.623**	
p,p' -DDT	<DL -0.091	<DL	0.014	0.032	14	0.002–0.147	0.024	0.042	0.054	20
p,p' -DDD	<DL-0.088	<DL	0.009	0.028	12	<DL-0.207	0.015	0.034	0.063	24
p,p' -DDE	0.004–0.286	0.025	0.070	0.105	47	<DL-0.215	0.018	0.062	0.084	28
ΣDDT	**0.004–0.465**	**0.025**	**0.093**	**0.165**		**0.002–0.568**	**0.057**	**0.138**	**0.201**	
t-Chlordane	0.010–0.345	0.008	0.086	0.116	53	0.001–0.126	0.015	0.036	0.044	34
c-Chlordane	<DL-0.141	<DL	0.018	0.046	16	<DL-0.172	0.002	0.036	0.070	19
ΣChlordane	**0.010–0.486**	**0.008**	**0.104**	**0.162**		**0.001–0.298**	**0.017**	**0.072**	**0.113**	
Methoxychlor	**<DL-0.034**	**0.003**	**0.007**	**0.013**	19	**<DL-0.066**	**0.001**	**0.015**	**0.024**	22
α-Endosulfan	<DL-0.033	<DL	0.005	0.012	12	<DL-0.027	<DL	0.004	0.010	9
β-Endosulfan	<DL-0.051	0.004	0.008	0.018	9	<DL-0.117	0.012	0.025	0.041	20
ΣEndosulfan	**<DL-0.084**	**0.004**	**0.013**	**0.030**		**<DL-0.144**	**0.012**	**0.029**	**0.050**	
Endosulfan Sulfate	**<DL-0.028**	**<DL**	**0.007**	**0.013**	12	**<DL-0.057**	**0.005**	**0.013**	**0.021**	24
ΣOCPs	**0.126–10.402**	**1.433**	**2.482**	**3.589**		**0.053–3.010**	**0.399**	**0.765**	**1.033**	

<DL = below detection limit.

Table 3
Comparison of the present results with other studies on the Ganga.

Sampling Time	Zone	Unit	Σ-HCH	Σ-DDT	Σ-ES	Reference
2014–2016	LZ	ng/mL	ND-2.940(↑)	ND - 1.311(↓)		Mondal et al. (2018)
Jun 2012	LZ	ng/L	ND- 114(↓)	ND - 5(↓)	ND - 10(↓)	Chakraborty et al. (2016)
2013–2014	UMZ1	μg/L	BDL-24.5(↑)	BDL-20.6(↑)	BDL(↓)	Raghuvanshi et al. (2014)
	UMZ2		BDL-13.6(↑)	BDL-18.6(↑)	BDL(↓)	
Dec 2010–Jan 2011	UZ	ng/L	5.5–7.74(↓)	ND-1.01(↓)	ND-0.92(↓)	Mutiyar and Mittal (2013)
Jun–Jul 2011	UMZ1		0.1–1(↓)	0.05–0.2(↓)	0.8–31.6(↑)	
Jun–Jul 2011	UMZ2		0.2–3.5(↓)	0.08–2.21(↓)	ND-85.4(↓)	
Jul–Aug 2011	LMZ		0.1–17.6(↓)	ND-12.3(↓)	ND-85.4(↑)	
2007	LMZ	ng/L	ND (summer)(↓) ND-48.3 (monsoon)(↓)		ND-157.3(↑)	Singh et al. (2012)
2008	LZ	μg/L	ND-6.65 (except lindane)(↓)	0.01–0.65(↑)		Ghose et al. (2009)
2003 (June to December)	UMZ1	μg/L	ND-0.260	ND(↓)		Shankararamakrishnan et al. (2005)
March 1989 and March 1990	UMZ1	μg/L	ND-1.38(↑)	N.D.(↓)	ND(↓)	Rehana et al. (1996)
March 1989 and March 1990	UMZ1	μg/L	ND-3.01(↑)	0.88–5.33(↑)	ND-0.75(↑)	Rehana et al. (1995)
August1992 (monsoon)	UMZ2	μg/L	ND-36.354(↑)	ND-79.818(↑)	ND-48.828(↑)	Nayak et al. (1995)

(↑) = Values higher than present study.
(↓) = Values lower than present study.
ND-Not Detected; BDL- Below Detection Limit – The values are reported as mentioned in the original paper/study.

considerably reduce the volume of water and concentrate the OCPs in the dry season. Additionally, the tributaries joining the Ganga in this zone, particularly the Ramganga and Kali, also bring significant domestic, industrial, and agricultural pollution loads, thereby increasing the pesticide burden in this zone (CPCB, 2016).

The lowest pesticide pollution gradient in the LMZ representing the states of Bihar and Jharkhand, in both seasons could be explained by the high drainage from major tributaries like the Ghaghra (94.4 billion cumecs), Gandak (52.2 billion cumecs), and Son (22.42 billion cumecs). These tributaries contribute significantly to the environmental flow (~5 times high as compared to other zones) and increased dilution factor (Table S9). In addition, the cropping intensity in this catchment is also low (Table 1) compared to that in other zones.

However, statistical analysis for difference between the zones was non-significant ($p > 0.05$) in both post-monsoon (*KW test, H (4),* $p = 0.34$) as well as post-winter (*KW test, H (4),* $p = 0.34$) seasons.

The non-significant difference among the zones could be attributed to the large number of point-sources (such as municipal and industrial waste water treatment drains) distributed all along the Ganga. A total of 139 drains carrying industrial and domestic sewage effluent join the Ganga directly. The storm water drains designed to flood-out the storm water during rainy season, are used for disposal of sewage and trade effluents, which ultimately joins the Ganga. These drains discharge pollution load of 361.2 tons per day (CPCB, 2016a) into the Ganga, having presence of pesticides that they may receive from the catchment area. A recent report from CPCB indicates that most of the drains joining Ganga had presence of organochlorine pesticides (CPCB, 2016b).

These factors may have eventually lead to the statistically non-significant difference between the zones despite the highest concentration of pesticide in the LZ and UMZ1.

3.2.1. Group wise abundance of OCPs in each zone

When assessing the zones for most abundant group of pesticides (Table S7), it was revealed that HCH isomers were found to be dominant (highest conc. and detection) in all zones for both seasons; however, in the dry season (post-winter) presence of other OCPs in also highlighted due to low dilution (Figs. 3b & 4).

The distribution of ΣHCHs across zones showed the order $\mathrm{LZ} > \mathrm{UMZ1} > \mathrm{UMZ2} > \mathrm{UZ} > \mathrm{LMZ}$ in the post-monsoon season and $\mathrm{UMZ1} \simeq \mathrm{LZ} > \mathrm{UMZ2} > \mathrm{UZ} > \mathrm{LMZ}$ in the post-winter season. The zone-wise statistical comparison (*KW test, H (4),* $p = 0.05$), in post-

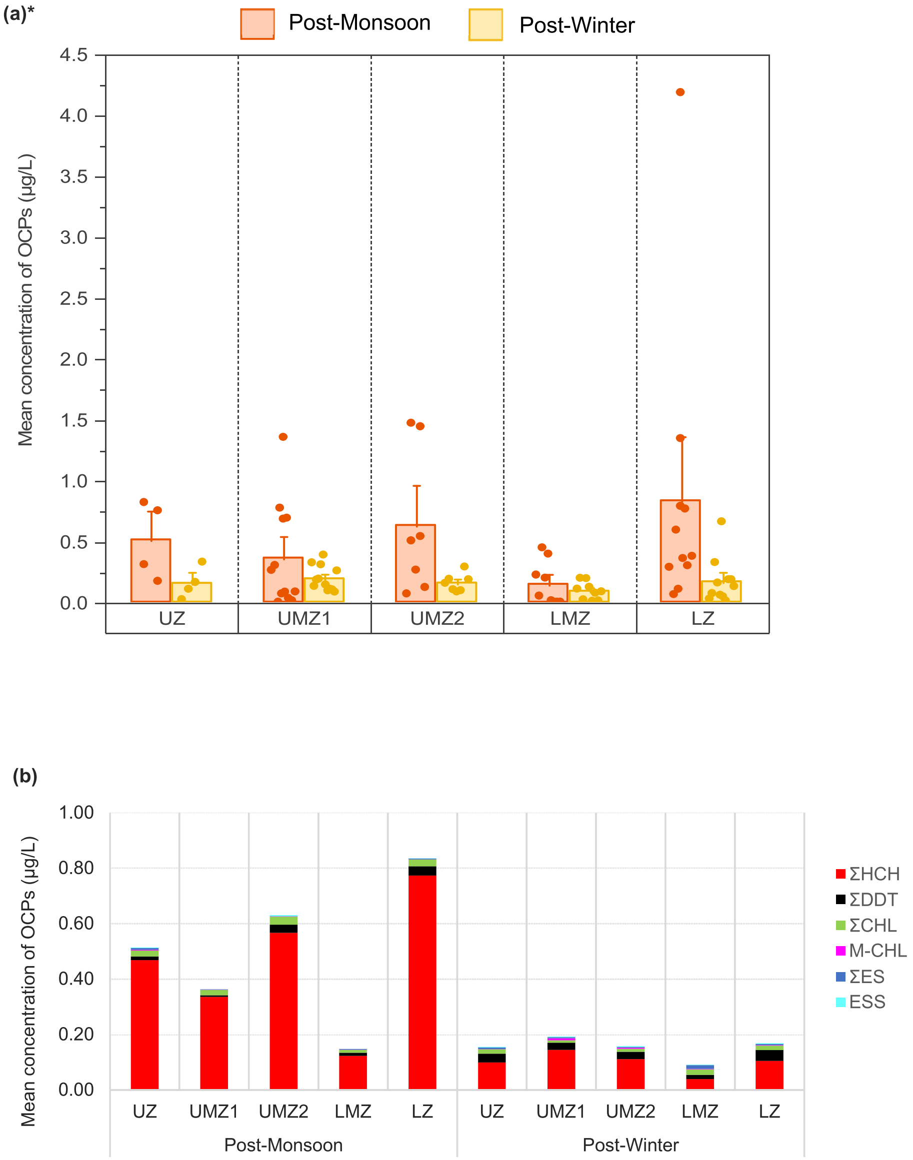

Fig. 3. Seasonal variation and zone-wise distribution of (a) total OCPs (b) Group-wise OCPs in Ganga River. *The bar represents mean Σ OCP and circles show conc. at each site in that zone.

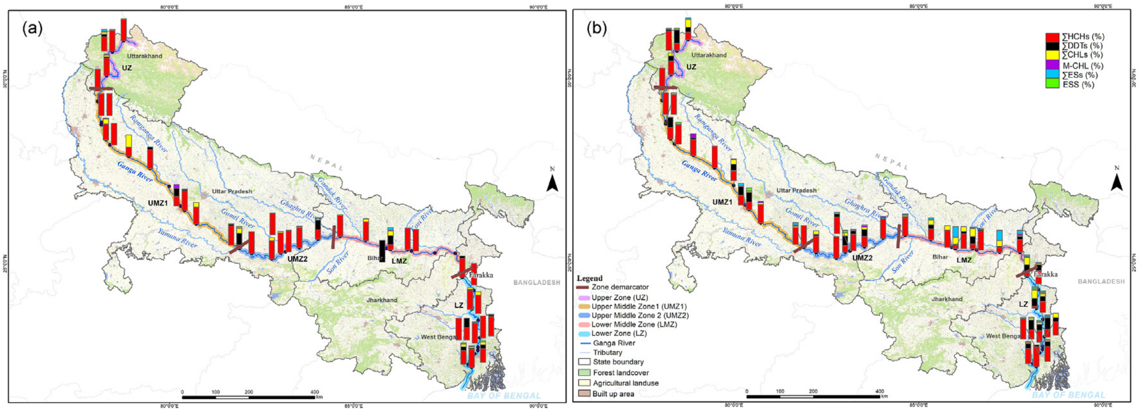

Fig. 4. Relative seasonal and spatial distribution of total OCPs in surface water samples of Ganga River in (a) Post-Monsoon season (b) Post-Winter season.

monsoon revealed no significant differences ($p > 0.05$) in ΣHCHs concentrations. However, significant differences (*KW test, H (4),* $p = 0.02$) were observed between UMZ1 and LMZ in the post-winter season.

For rest of the zones the difference was statistically insignificant. The environmental flow and contribution of tributaries were considered as major factors influencing the significant difference, in ΣHCHs concentrations, between UMZ1 and LMZ. In the UMZ1, a substantial amount of water is diverted from the river to support agricultural activities through a system of canals, which may considerably reduce the volume of water and concentrate the OCPs in the dry season (post-winter season). Further, the small tributaries joining the Ganga in this zone, particularly the Ramganga and Kali, also bring significant domestic, industrial, and agricultural pollution loads, thereby increasing the pesticide burden in this zone (CPCB, 2016a). In addition, in a recent study from CPCB, a large number of drains on the Ganga (17 out of 33) were found to contain HCHs (range 0.1–7.03 ng/l) as the dominant pesticide. Whereas in the LMZ, the factors of high environmental flow and low cropping intensity, as mentioned in the earlier section, are the contributing factors for lower concentration of ΣHCHs.

The ΣDDT distribution among the five zones followed the order LZ > UMZ2 > LMZ > UMZ1 > UZ in the post-monsoon season (*KW test, H (4), p = 0.10*) and LZ > UMZ1 > UMZ2 > LMZ > UZ in the post-winter season (*KW test, H (4), p = 0.21*). Comparison of the mean concentration of ΣCHLs among the five zones revealed the distribution pattern as LZ > UMZ1 > UMZ2 > LMZ > UZ in the post-monsoon season (*KW test, H (4), p = 0.24*) and LMZ ≃ LZ > UMZ1 > UMZ2 > UZ in the post-winter season (*KW test, H (4), p = 0.62*). The spatial distribution pattern of ΣESs was LZ > LMZ > UZ > UMZ1 > UMZ2 in the post-monsoon season (*KW test, H (4), p = 0.41*) and LMZ > LZ ≫ UMZ1 > UZ ≃ UMZ2 in the post-winter season (*KW test, H (4), p = 0.23*). For M-CHLR, the distribution in the post-monsoon (*KW test, H (4), p = 0.37*) and post-winter season (*KW test, H (4), p = 0.67*) was in the order UZ > UMZ1 > LMZ > UMZ2 > LZ and LMZ > LZ > UMZ1 > UZ ≃ UMZ2, respectively. For the ESS distribution trend in the post-monsoon season (*KW test, H (4), p = 0.20*) was UMZ2 > LZ > UZ and in the post-winter season (*KW test, H (4), p = 0.47*) was LZ > UMZ1 > LMZ > UMZ2 > UZ. However, the zone-wise statistical comparison for both seasons revealed no significant differences ($p > 0.05$) in the concentrations of any of the above pesticide groups.

3.3. *Source apportionment of OCPs by ratio analysis*

The ratio diagnostic method has been extensively used in the past to identify the potential sources of OCP contamination (Syed and Malik, 2011; Mahmood et al., 2014; Sultana et al., 2014; Yu et al., 2014; Baqar et al., 2018). All the target OCPs investigated in this study are either banned or restricted. Therefore, it is important to ascertain the source (ongoing or historic) of these pesticides. Composition differences between target analytes in surface water environment provide useful information to determine the different pollution sources.

3.3.1. *HCHs*

Technical-grade HCHs contain a mixture of 60%–70% α-HCH, 5%–12% β-HCH, 10%–15% γ-HCH, 6%–10% δ-HCH and 3%–4% ε-HCH, whereas technical-grade lindane contains ~99% γ-HCH (Iwata et al., 1993; Qiu et al., 2004; Gao et al., 2013; Yu et al., 2014; Mahmood et al., 2014). To identify the potential source of HCH contamination, the α-HCH/γ-HCH ratio is used. In general, the α-HCH/γ-HCH ratio in technical-grade HCHs usually ranges between 3 and 7. A ratio value < 3 indicates fresh inputs of γ-HCH. An α-HCH/γ-HCH ratio >7 indicates long-range transport or the photochemical breakdown of γ-HCH into α-HCH (Barrie et al., 1992; Willett et al., 1998; Wang et al., 2018). The α-HCH/γ-HCH ratio in our study was found to be < 3 in all five zones for both seasons, thus indicating the possible ongoing illegal usage of HCH in the study area. Additionally, relatively low prevalence of β-HCH (Law et al., 2001; Tan et al., 2009), the most persistent HCH isomer, also indicates fresh inputs of HCH (Fig. 5a). We anticipate that the reason for the detection of fresh inputs of technical-grade HCH (particularly lindane) could be linked to their application in paddy fields. In the UMZ1, UMZ2, LMZ, and LZ, rice is generally grown under rainfed conditions along the Ganga basin; these zones (and the corresponding states) are the top cultivators/producers of rice in the country. The elevated concentrations of HCH detected during the wet season in this study indicates that this insecticide is largely applied during the flowering season of paddy (Ramesh et al., 1991).

3.3.2. *DDTs*

Technical-grade DDT usually contains 77.1% *p,p*′-DDT, 14.9% *o,p*′-DDT, and 4% *p,p*′-DDE (Yu et al., 2014; Baqar et al., 2018). However, with time, under favorable aerobic and anaerobic conditions, the parent compound DDT breakdowns to its metabolites DDE and DDD respectively (Heberer and Dünnbier, 1999; Hitch and Day, 1992). Therefore, the ratios of DDD/DDE could be used to indicate the aerobic (DDD/DDE < 1) or anaerobic (DDD/DDE > 1) degradation (DDD/DDE>1) of DDT. Similarly, the ratio of DDT/(DDE + DDD) is generally used to estimate the age of DDTs; with time, DDT will degrade into DDE and DDD, resulting in a gradual decrease in the level of DDT and subsequent increase in the levels of DDE and DDD (Harner et al., 1999). Therefore, a ratio of DDT/(DDD + DDE) < 1 indicates past usage and historic contamination, whereas a ratio > 1 indicates that there are fresh inputs of DDTs (Gao et al., 2013; Wang et al., 2018). For this study, the ratios of DDD/DDE and DDT/(DDE + DDD) are shown in Fig. 5b. As shown in the figure, at the majority of the sites, the ratio of DDD/DDE was <1, indicating that biodegradation of DDT occurs predominantly under aerobic conditions. Furthermore, in this study, the calculated ratio of DDT/(DDD + DDE) was <1 at the majority of sites (in all zones) for both seasons, indicating that DDT contamination in the Ganga was mainly from its metabolites and historic usage. However, during the post-winter season, the ratio analysis revealed fresh inputs of DDTs at some sampling points in the UZ, UMZ1, LMZ, and LZ. As stated in the previous section (Section 4.2), this could be because of the current use of DDT for the control of malaria vectors under the public health program.

3.3.3. *Chlordane*

Technical-grade chlordane consists of the major components t-CHL (13%), c-CHL (11%), trans-nonachlor (5%), heptachlor (5%), and over 30 less abundant chlordanes, nonachlors, and chlordenes (Bidleman et al., 2002). Trans-chlordane, owing to its lower half-life (Table S1), degrades relatively faster in the environment than its cis counterpart (Yamada et al., 2008). Therefore, a ratio of cis-chlordane to trans-chlordane < 1 specifies the fresh usage of chlordane (Bidleman et al., 2002; Yu et al., 2014). In the current study (Fig. 5 c), the ratios calculated for cis/trans-chlordane were <1 at all sampling zones, indicating the ongoing usage of chlordane in the study area.

3.3.4. *Endosulfan*

The endosulfan isomers α-endosulfan, and β-endosulfan constitute up to 70% and 30% of the composition of the technical-grade endosulfan mixture, with the α-isomer being relatively more degradable (Table S1) in the environment than the β-endosulfan (Jiang et al., 2009). The ratio of α-endosulfan/β-endosulfan is used to calculate the age and source of endosulfan in the

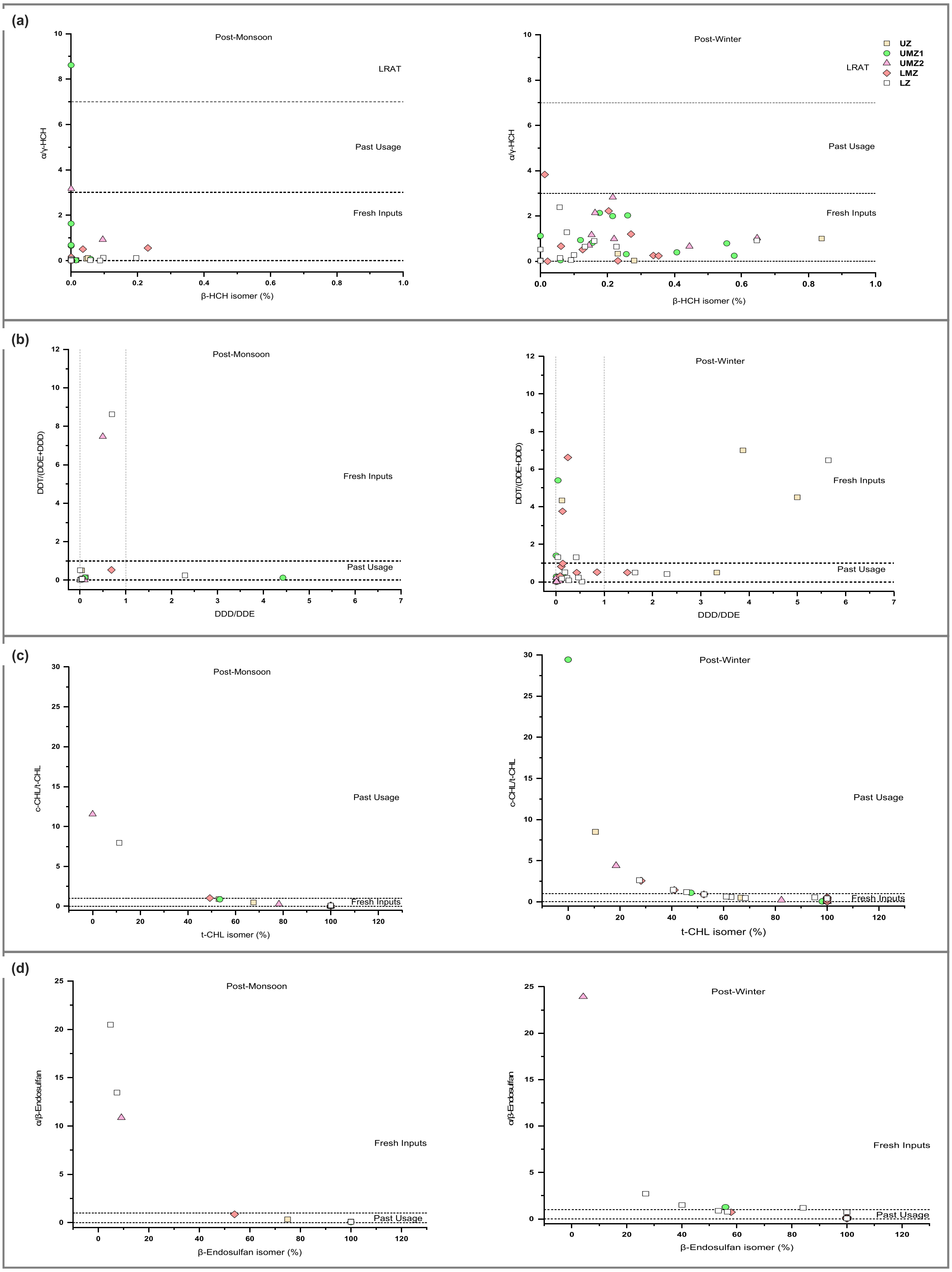

Fig. 5. Source apportionment analysis of OCPs for Post-Monsoon and Post-Winter (a) HCH (b) DDT (c) Chlordane (CHL) (d) Endosulfan (ES).

environment. In general, a value < 1 indicates past usage (Yu et al., 2014). In our study, the ratios of α-endosulfan/β-endosulfan at almost all zones for both seasons were found to be < 1, indicating past usage in these areas (Fig. 5d). However, at some sampling sites in the UMZ2 (post-winter) and LZ (post-monsoon), the ratios were >1, indicating fresh inputs of technical-grade endosulfan.

4. Comparative assessment of present results with other rivers

4.1. Comparison with other studies on the Ganga

To assess trends and patterns regarding OCPs in the Ganga, the results of this study were compared against OCP contamination data from previous studies on the Ganga (Table 3). A limited number of studies were available on OCP contamination in the Ganga from the past two decades, and the majority of these focused on specific small stretches of the river, giving valuable yet incomplete information on the OCP contamination status of river as a whole. In addition, data on temporal trends of OCPs in the Ganga are also very limited. The OCP levels reported in this study point toward a mixed trend where some studies showed the OCP contamination lower than our study while a few reported higher than this study. The ∑HCHs in our study was lower than 60% of the previous studies, whereas for ∑DDTs and ∑ESs, we found that our results were lower than 50% of the previous studies. The results highlight both the success and failure of the government's efforts to ban the target OCPs in India. After the ratification of the Stockholm Convention in 2006, India executed its first National Implementation Plan (NIP) in 2011, wherein the government developed its strategy to deal with these chemicals and subsequently implemented regulations. However, there has been substantial delay in implementing the initiatives to mitigate POPs in the country. The results of our study highlight that the situation has improved compared to that in the past; however, to see 100% success and a complete downward trend, a stricter implementation of the NIP, as well as more time is required to observe a greater reduction in OCP distribution.

4.2. Comparisons with other Indian and global rivers

When comparing the results of present study to similar studies on other Indian rivers, we observed that in almost all studies, with the exception of a few, the levels of target pesticides were lower than the levels reported in this study (Table S11). As with any large river, the issues of constructing dams and barrages are known. Additionally, a large number of industrial and sewage drains also release toxic wastes into the water. The Ganga has three major dams and six barrages, which reduces >80% of its flow till it completes half of its journey into the UMZ1. Afterwards, the additional water input from its tributaries does help to dilute the pollution load for a few hundred kilometers, but a major barrage in the LZ (Farrakka) again diverts substantial water to Bangladesh. The water is also abstracted through several feeder canals and diverted for local irrigation needs. Further the Ganga has 139 drains and 767 grossly polluting industries releasing its wastes (both treated and untreated) into the already flow-stressed river, thereby increasing the pollution load (CPCB, 2016a, b). Finally, the use of a considerably large catchment area of the Ganga (compared to other small rivers) for agriculture adds high quantities of pesticides to the river. Hence, the higher concentration of OCPs reported in the Ganga compared to that in other Indian rivers is valid given its extent and other reasons specified above.

Similarly, we compared the results from this study with studies on OCP contamination in global freshwater bodies from Asia, Africa, and developed nations. It was observed that barring few studies, most of the rivers had lower levels of ΣHCHs, ΣDDTs, and ΣCHL than our study, pointing toward the late ban and poor implementation of the regulations in India. A summary of the comparison is presented in Table S12.

5. Potential ecological risk assessment

For this study, we compared the distribution of OCPs in the surface water of the Ganga against the guideline values (GVs) established by USEPA (Table S15). We found that the concentrations for most of the pesticide groups exceeded these GVs for aquatic life criteria at multiple sites across all the zones. The concentrations of ΣHCHs were above the USEPA GV at 12% sites in post monsoon whereas in post-winter it was within the permissible limits at all the sites. Similarly, ΣDDTs exceeded maximum criteria threshold at 51% sites in post-monsoon and 72% sites in post-winter. For ΣCHL, the concentrations were observed above the GVs at 56% sites in post-monsoon and 51% sites in post-winter. The concentrations of methoxychlor were higher than the GVs at 2% sites in both the seasons. For ΣESs, all the sites were within the permissible GVs in both the seasons.

Further, we calculated the risk quotients (RQs) associated with exposure to individual OCPs for three representative trophic levels, viz., algae, aquatic invertebrates, and fish. To identify the high-risk zones, the mean and maximum detected concentrations at each zone were used to determine the general (RQm) and worst-case (RQex) scenarios, respectively. The PNECs, RQs and values corresponding to the maximum and mean concentrations of individual OCPs are presented in Tables S13, S14, and S15 and Fig. 6, respectively.

The RQm values derived from the seasonal data revealed a high ecological risk in the LZ, whereas the RQex values showed ecological risks at the majority of the zones. Among all pesticides, p,p′-DDE showed a $RQ > 1$ at all monitored sites followed by β-endosulfan, p,p′-DDT and p,p′-DDD, which also showed high ecological risks at the majority of sites, particularly in the post-winter season. The high risk posed by these contaminants is mainly because of their relatively high toxicity to fish, algae, or aquatic invertebrates, hence producing quite low PNEC values. Likewise, δ-HCH showed a high risk in the LZ whereas α-ES showed a high risk in the UMZ2, LMZ, and LZ in both seasons. The ecotoxicological risk assessment highlights that these pesticides ($RQ > 1$) should be prioritized for risk management, particularly because the Ganga is also home to diverse fauna and endangered species. High and continuous exposure to OCPs can compromise the normal function of key physiological processes, which can ultimately affect the survival of these aquatic species.

Furthermore, chlordane (trans and cis), methoxychlor, and γ-HCH showed medium to negligible risks in all zones in both seasons. However, we suggest that the estimated ecological risks should not be overlooked in the current scenario, considering the high concentration of these pesticides detected in the LZ of the Ganga, and their ability to pose negative impacts on aquatic ecosystems.

In the long term, the high aquatic risk caused by pesticides may lead to changes in the fish and invertebrate communities, leading to decreases in the most sensitive species and increases in the most resistant ones, with consequent loss of biodiversity (Palma et al., 2014; Kuzmanović et al., 2015; Zhang et al., 2016).

6. Conclusion

Thirteen OCPs, their source, and the ecological risk were investigated in surface water of Ganga along 43 sites for two seasons for the first time. A significant difference ($p < 0.05$) in OCP concentrations was found between wet and dry seasons. γ-HCH (lindane) was the most frequently detected compound and found to have fresh inputs across all the zones. Another pesticide chlordane also had fresh inputs across all the zones. The results also point toward the consequences of continued use of DDT in health programs in India. Long-range atmospheric transport and fresh inputs are factors that contribute toward high levels of HCH and DDT in the relatively pristine UZ and are of serious concern. Though different zones showed high concentration of pesticides during different seasons, however the statistical difference amongst the zones was insignificant indicating the contribution of point source pollution from the open drains. The ecotoxicology risk assessment showed that the concentrations of most of the OCPs exceeded international GVs for the protection of aquatic life at several sites. A

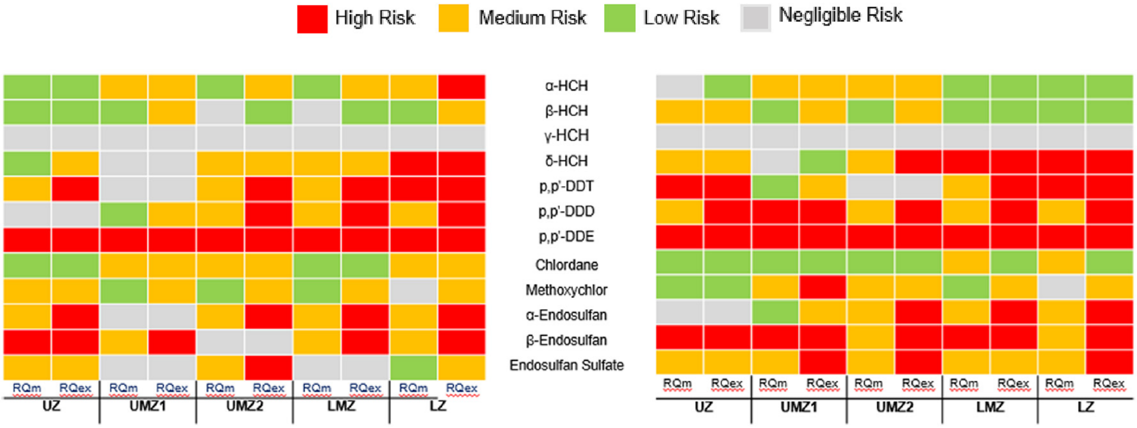

Fig. 6. RQm and RQex of individual OCPs in surface water of the Ganga River (a) Post-Monsoon (b) Post-Winter.

possible high risk to aquatic biota from DDE, DDT, δ-HCH, and α-ES at all or most of the sites was pointed out by RQ model. Pesticides in aquatic ecosystems usually occur as mixture of multiple pesticides than individually, therefore, further research on the potential combined eco-toxicological effects (synergistic, additive, or antagonistic) of OCPs mixture is required.

This study highlights that the Ganga is subjected to contamination by banned and restricted OCPs, and spatial variations have indicated different sources, hydrological differences in the system, and different land-use patterns. Our study validated our hypothesis that there is continued use of banned OCPs at a sizeable spatial level in the Ganga states; this poses a potential risk to the aquatic biodiversity of the river. Since this is the first study to include the spatial and temporal coverage along the length of the river, the present findings could be effectively used in devising a state-wise policy and measures to tackle the illegal usage of OCPs. High levels of OCPs in the post-winter season emphasize the need to maintain minimum environmental flow in the river that is currently compromised due to water abstraction and construction of several dams and barrages on the Ganga. Further, monitoring of drains along the river need to be taken up for the presence of pesticide concentrations and loads. In light of the results presented in this study, we suggest that awareness campaigns and training programs for farmers must be taken up on a priority basis to eliminate the use of banned pesticides. Besides, adoption of integrated pest management, mixed intercropping systems, and organic agroforestry systems must be encouraged. Stricter implementations of the NIP on POPs and timely updates for the next phase is also recommended. We also propose a continuous monitoring campaign to follow up on the occurrence of pesticide residues in the Ganga. Finally, to ensure a healthy ecosystem for the biodiversity of the Ganga, we advocate a holistic ecological risk assessment that includes population-level risk assessment at each critical habitat to evaluate the long-term effects of multiple stressors on aquatic populations.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

CRediT authorship contribution statement

Ruchika Sah: Conceptualization, Methodology, Formal analysis, Investigation, Data curation, Writing - original draft. **Anju Baroth:** Conceptualization, Validation, Writing - original draft, Writing - review & editing, Visualization, Supervision. **Syed Ainul Hussain:** Resources, Supervision, Funding acquisition.

Acknowledgement

We thank National Mission for Clean Ganga, Ministry of Jal Shakti, Department of Water Resources, River development and Ganga Rejuvenation, Government of India for funding and supporting this research (Grant No. B-02/2015-16/1259/NMCG-WII-PROPOSAL). Authors are thankful to Director and Dean, Wildlife Institute of India for their support. We thank Dr J A Johnson for his support and help as the component coordinator and as Research Lab Incharge. Mr. Chetan PS Ahada, Ms. Sunanda Bhola, Ms. Pooja Chaudhary and the staff of Ecotoxicology Lab, WII is thanked for their critical support in the field and lab. Ms. Aishwarya Ramachandran is thanked for her assistance and guidance in GIS mapping. We would also like to thank Prof. Qamar Qureshi, Praveen Krishna and Nilanjan Chatterjee for their help in statistics.

Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.envpol.2020.114229.

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