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Research Paper

Legacy and new chlorinated persistent organic pollutants in the rivers of south India: Occurrences, sources, variations before and after the outbreak of the COVID-19 pandemic

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ABSTRACT

During pre-pandemic time, organochlorine pesticides (OCPs) and polychlorinated biphenyls (PCBs) were investigated in the surface water of Periyar River (PR) and Bharathappuzha River (BR) in Ernakulam and Malappuram districts of Kerala, respectively and Adyar River (AR) and Cooum River (CR) in Chennai district of Tamil Nadu. After the outbreak of COVID-19 pandemic, variation in OCPs and PCBs were evaluated for AR and CR. Dominance of $\beta$-HCH and $\gamma$-HCH in south Indian rivers indicate historical use of technical HCH and ongoing use of Lindane, respectively. In $>90\%$ sites, $p,p'$-DDT/$p,p'$-DDE ratio was $<1$, indicating past DDT usage. However during the outbreak of the COVID-19 pandemic, elevated $p,p'$-DDT in AR and CR reflects localized use of DDT possibly for vector control. Similarly, during the first wave of pandemic, over a 100-fold increase in PCB–52 in these rivers of Chennai mostly via surface run-off and atmospheric deposition can be reasoned with open burning of dumped waste including added waste plastic in the solid waste stream. On contrary, a significant ($p<0.05$) decline of dioxin-like PCBs level, suggests lesser combustion related activities by the formal and informal industrial sectors after the lockdown phase in Tamil Nadu. Eco-toxicological risk assessment indicated a higher risk for edible fish in PR due to endosulfan.

1. Introduction

Legacy persistent organic pollutants (POPs) such as organochlorine pesticides (OCPs) and polychlorinated biphenyls (PCBs) are of major concern for public health owing to their persistence, carcinogenicity, bioaccumulation, and the ability for long-range atmospheric transport...
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2. Materials and method
2.1. Study area
In Kerala, Periyar River (PR) is the longest perennial river and Bharathappuzha River (BR) forms the largest river basin. These rivers empty into the Arabian Sea along the south-west coast of India. They are the main source of freshwater for about 7 million inhabitants from Ernakulam and Malappuram districts. Adyar River (AR) and Cooum River (CR) are flowing in the Chennai district (population: 11.2 million) of Tamil Nadu along the south-east coast of India. These rivers were once a prominent freshwater source in Chennai but now function as the wastewater carrier for the city, draining into the Bay of Bengal. A total of forty composite surface water samples were collected from these four rivers during August-September 2019. Due to the outbreak of the COVID-19 pandemic in the year 2020 and the implementation of travel restrictions, samples during the pandemic (August-September 2020) were collected only from Chennai. Details of the sampling sites are given in Fig. 1, and Table S1. Composite water samples were collected according to the procedure mentioned elsewhere (Chakraborty et al., 2016a). Briefly, from each site, composite water sample of 1 L was prepared after mixing 5 samples of 1 L each over a length of 500 m. Samples were transferred into pre-cleaned amber tarsion bottles, labelled, and transported to the laboratory in an icebox. The samples were stored at \( -20 \degree C \) until extraction.

2.2. Extraction of PCBs and OCPs
Water samples were extracted based on the method used for chlorinated pesticidal POPs in a previous study (Khumar and Chakraborty, 2019). Briefly, surface water samples were filtered through 0.45 \( \mu \)m pore size and extracted by solid-phase extraction. A mass of 20 ng of 2,4,5-trichlorophenol, 2,4,6-trichlorophenol, pentachlorophenol (PCP), hexachlorophene, decachlorobiphenyl, and dieldrin were added to each sample before extraction to estimate the recovery percentage of two categories of chlorinated POPs. The extracts were concentrated to complete dryness and solvent-exchanged to n-hexane (1–2 mL). Further, the extracts were purified using an alumina/silica column. Eluates were reduced to 0.5 mL using a gentle stream of nitrogen before quantification.

2.3. Instrumental analysis
OCPs and PCBs were analyzed using a gas chromatograph (GC) (Agilent 7890B) equipped with an electron capture detector (ECD) using HPS column (30 m, 0.32 mm \( \times \) 0.25 \( \mu \)m). Nitrogen was used as the carrier gas with a total flow of 18.845 mL/min and the detector temperature was 290 \degree C. The injection volume was fixed as 1 \( \mu L \) and the samples were injected in splitless mode. Prior to analysis, 20 ng of pentachloronitrobenzene (PCNB) was added to each extracted sample as the internal standard. Twenty OCPs viz., \( p,p'-DDE, p,p'-DDD, p,p'-DDT, \) \( \alpha\text{-HCH, } \beta\text{-HCH, } \gamma\text{-HCH, } \delta\text{-HCH, } \alpha\text{-endosulfan, } \beta\text{-endosulfan, endosulfan sulfate, cis-chlordane, trans-chlordane, aldrin, } \) \( \delta\text{-dieldrin, } \alpha\text{-dieldrin, } \beta\text{-dieldrin, } \gamma\text{-dieldrin, } \) \( \alpha\text{-endrin, } \beta\text{-endrin, } \gamma\text{-endrin, } \) heptachlor, heptachlor epoxide and methoxychlor, were quantified. A total of twenty five PCB congeners were analysed. The list of PCB congeners include dl-PCB (\(-8\)), tri-PCBs \(-18, -28\), tetra-PCBs \(-44, -52, -70, -77, -81\), penta-PCBs \(-101, -105, -114, -118, -123, -126\), hexa-PCBs \(-138, -151, -153, -156, -157, -167, -169\), hepta-PCBs \(-180, -189\), and octa-PCBs \(-194, -195\). For OCPs, the initial temperature of 120 \degree C was ramped to a final temperature of 290 \degree C with a hold time of 3.5 mins. For PCBs, the initial oven temperature was maintained at 150 \degree C for an identification of sources using diagnostic ratios and compositional profiles, (iii) variation in OCPs and PCBs, before and after the outbreak of COVID-19 pandemic in rivers of Chennai, (iv) eco-toxicological risk due to the pesticidal POPs, and (v) toxic equivalents (TEQs) due to dl-PCBs.
equilibration time of 1 min (3 mins hold time) and was increased to 290 °C at a rate of 5 °C/min (5 mins hold time).

2.4. QA/QC

Co-planar isotope labelled PCB-Mix and indicator PCBs and dl-PCBs standards were procured from Wellington Laboratories. PCB-209, TCmX and OCP-Mix were purchased from Sigma Aldrich, USA. One procedural blank was run for every 7 samples, and multi-level calibration curves and calibration verifications ($R^2 \geq 0.999$) were conducted for OCPs and PCBs. Signal to noise ratio was greater than 3. Limits of detection (LOD) for OCPs and PCBs are given in Table S2 and S3, respectively. LOD for OCPs and PCBs varied between 0.001 and 0.072 ng/L and 0.01–0.04 ng/L, respectively. Concentrations lower than LODs were reported as non-detected for quantified OCPs and PCBs. Surrogate recovery for TCmX and PCB-209 were 88–117 % and 85–110 %, respectively. For isotope labelled co-planar PCBs, the recovery ranged between 81 % and 117 %.

2.5. Annual flux estimation

The annual fluxes of OCPs and PCBs in the perennial rivers of Kerala (BR and PR) flowing into the Arabian Sea was calculated by the following formula given elsewhere (Eqani et al., 2012):

$$F = C \times R \times 10^{-12}$$

where the flux is denoted by $F$ (tons/year), $C$ is the mean environmental concentration (ng/L) in the river, and $R$ is the annual flow rate (m$^3$) of the river. AR and CR are more stagnant rivers, and their flow rates are unavailable. The average annual flow for BR and PR were 5082.9 million m$^3$ and 4867.9 million m$^3$, respectively (ENVIS, 2022).

2.6. Eco-toxicological risk assessment

Eco-toxicological risks for OCPs were estimated by calculating the hazard quotient (HQ) based on USEPA guidelines (USEPA, 1998) given by the formula.

$$HQ = \frac{EC}{PNEC}$$
where, EC is the environmental concentration at each site and PNEC is the predicted no-effect concentration of aquatic species. PNEC values for each organism are given elsewhere (Chakraborty et al., 2016a).

2.7. Toxic equivalents

Toxic equivalents (TEQs) were estimated for twelve dl-PCBs including the mono-ortho (PCB–105, –114, –118, –123, –156, –157, –167, –189), and non-ortho congeners (PCB–77, –81, –126, –169) by using the corresponding Toxic Equivalency Factors (TEFs) provided by the World Health Organization (Van den Berg et al., 2006).

2.8. Statistical analysis

All statistical analyses, including box-whisker plots, correlation analysis, t-test, ANOVA, and principal component analysis (PCA) were performed using IBM SPSS 22.0 windows version.

3. Results and discussion

3.1. Spatial distribution and sources

The range of each OCP and their spatial distribution in the four river networks of south India are given in Table 1 and Fig. 1, respectively. Range of each PCB congener for all the surface water samples are given in Table 2. In general, DDT showed maximum detection frequency (75 %), followed by HCH (55 %) and endosulfan (40 %). The detection frequency of pesticides was least for Adyar River. On contrary, detection frequency for indicator PCB congeners varied between 8 % and 60 % frequency of pesticides was least for Adyar River. On contrary, detection frequency for tri and tetra PCB homologues. It is to be noted that Kerala experienced extremely heavy rainfall during the sampling time (August 2019) leading to intense flooding in major districts of Kerala. In 2020, Chennai received relatively higher rainfall than previous years due to north-east monsoon. Therefore surface run-off and atmospheric deposition are expected to be the major pathways for POPs in the rivers of south India.

### Table 1

| Compounds              | Bharathappuzha (n = 9) | Periyar (n = 9) | Cooum (n = 6) | Adyar (n = 3) |
|------------------------|------------------------|----------------|--------------|--------------|
| (ng/L)                 | Range (Avg ± SD)       | Range (Avg ± SD) | Range (Avg ± SD) | Range (Avg ± SD) |
| α-HCH                  | ND-1.41(0.61 ± 0.72)   | ND-1.34(0.29 ± 0.58) | ND-1.39(0.87 ± 0.68) | ND            |
| β-HCH                  | ND-6.03(2.64 ± 3.16)   | ND-5.95(2.62 ± 2.62) | ND-6.61(1.33 ± 1.88) | ND            |
| γ-HCH                  | ND-7.95(2.64 ± 3.96)   | ND-7.86(2.62 ± 2.62) | ND-7.95(2.44 ± 3.80) | ND-7.95(2.65 ± 4.59) |
| δ-HCH                  | ND                    | ND              | ND-0.15(0.04 ± 0.06) | ND            |
| Σ2HCH                  | ND-15.37(5.90 ± 7.42)  | ND-15.11(2.49 ± 5.12) | ND-12.75(4.12 ± 5.52) | ND-7.95(2.65 ± 4.59) |
| p,p’-DDE               | ND-7.15(1.14 ± 2.34)   | ND-2.77(0.45 ± 0.91) | ND-5.99(1.99 ± 2.40) | 0.39-3.00(1.79 ± 1.31) |
| p,p’-DDD               | ND                    | ND              | ND            | ND            |
| p,p’-DDE               | ND                    | ND              | ND            | ND            |
| ΣDDT                   | ND-7.15(1.14 ± 2.34)   | ND-7.19(1.87 ± 2.93) | ND-5.99(1.99 ± 2.40) | 0.39-3.00(1.79 ± 1.31) |
| α-Endosulfan           | ND                    | ND              | ND            | ND            |
| Endosulfan sulfate     | ND-4.13(0.46 ± 1.38)   | ND-4.10(0.46 ± 1.37) | ND-4.06(0.68 ± 1.66) | ND            |
| β-Endosulfan           | ND                    | ND              | ND            | ND            |
| Σ2ENDO                 | ND-4.13(0.46 ± 1.38)   | ND-7.21(2.16 ± 3.17) | ND-7.21(2.50 ± 2.21) | ND-4.06(0.68 ± 1.66) |
| cis-chlordane          | ND-2.93(0.33 ± 0.9)    | ND              | ND            | ND            |
| Trans-chlordane        | ND-2.69(0.30 ± 0.90)   | ND-2.55(0.28 ± 0.85) | ND              | ND            |
| ΣChlorodanes           | ND-5.62(0.62 ± 1.87)   | ND-2.35(0.28 ± 0.85) | ND              | ND            |
| Aldrin                 | ND-0.19(0.04 ± 0.18)   | ND              | ND-0.01(0.03 ± 0.04) | ND-0.06(0.02 ± 0.03) |
| Endrin                 | ND                    | ND              | ND            | ND            |
| Dieldrin               | ND-5.84(0.65 ± 1.95)   | ND              | ND            | ND            |
| Endrin aldehyde        | ND                    | ND              | ND            | ND            |
| Heptachlor             | ND                    | ND              | ND            | ND            |
| Heptachlor epoxide     | ND-4.40(1.34 ± 1.44)   | ND-2.44(1.04 ± 1.05) | ND-0.61(0.10 ± 0.25) | ND            |
| Methoxychlor           | ND-4.85(1.06 ± 2.11)   | ND-4.89(2.15 ± 2.55) | ND              | ND            |
| Endrin ketone          | ND-6.08(5.33 ± 2.00)   | ND-7.02(4.76 ± 2.72) | ND-6.37(5.02 ± 2.47) | ND-6.32(4.14 ± 3.59) |

(*ND-Not detected.)

### 3.1.1. Legacy and newly enlisted pesticidal POPs

3.1.1.1. DDT. In general the sum of DDT isomers and metabolites (ΣDDT=p,p’-DDE + p,p’-DDD + p,p’-DDT) in the rivers of south India showed a dominance of p,p’-DDE (53–86 %) thereby indicating the extensive use of technical DDT in the past. However in PR the dominance of p,p’-DDT (60 % of ΣDDT) reflects ongoing use of DDT. The highest concentration of ZDDT was seen in an upstream site of PR (PR-03, Thiruvalikunjam, 7.19 ng/L), with 80 % contribution from p,p’-DDT. It is to be noted that the upstream region of PR is surrounded by agricultural land known for growing rice as one of the major crop produced in Ernakulam district (CMFRI, 2022). Among 13 states from different geographical regions in India, the highest concentration of DDT was observed in rice samples of Kerala (10 mg/kg) due to the agricultural use of DDT (Toteja et al., 2003). Furthermore, elevated level of soil-borne p,p’-DDT in specific sites of Kerala has been earlier reasoned with the usage of technical DDT formulation (Khumar et al., 2020). Hence the possibility of agricultural use of DDT in Kerala cannot be ruled out. In CR and AR, p,p’-DDE was the only abundant metabolite. Unlike Kerala, the highest ΣDDT level was seen in Tamil Nadu with 100 % contribution from the most stable metabolite of DDT, p,p’-DDE at CR-03 (Pudupet, 5.99 ng/L) before the outbreak of the pandemic. The average concentration and dominance of p,p’-DDT in south Indian rivers were in line with levels reported from urban and suburban transects of the lower stretch of River Ganga (Khumar and Chakraborty, 2019) and Yongding River Basin, China (Wang et al., 2018).

The dominance of this stable DDT metabolite in Chennai might be a combined impact of atmospheric deposition (Chakraborty et al., 2010) and surface run-off from saturated contaminated soil (Chakraborty et al., 2015). In general, the average concentration of p,p’-DDT in rivers of south India seems to be lower than that of River Brahmaputra, and River Hooghly flowing through north-eastern and eastern part of India, respectively (Chakraborty et al., 2016a; Khuman and Chakraborty, 2019), and River Yamuna in northern India (Kausik et al., 2008; Kumar et al., 2012), River Chenab in Pakistan (Eqani et al., 2012), Talar, Babolrood and Haraz rivers in Iran (Behrooz et al., 2020) and Maozhou, Guanlan, Buji, Longgan and Pingshan rivers in China (Qiu et al., 2021), respectively.
but comparable with River Pangani in Tanzania (Hellar-Kihampa et al., 2013) (Table S5). It is to be noted that unlike Kerala, in the rivers of Tamil Nadu, p,p'-DDT was detected only during the pandemic with the highest p,p'-DDT evidenced at Koyambedu (CRC-2, 0.58 ng/L), indicating fresh use of DDT (Fig. 2 and Table S4).

### 3.1.1.2. HCH

In south Indian rivers, the highest concentrations of sum of HCH isomers (ΣHCH= α-HCH + β-HCH + γ-HCH + δ-HCH) in BR and PR was observed at Ponnani Harbor (BR-09, 15.37 ng/L) followed by Ponnani Pullimut; BR-7, Pambodi; BR-2, Thavanoor) and at a specific site in PR (PR-08, Moothakunnamb Harbor). Diagnostic ratio of α/γ-HCH in these sites were < 1 hence we suggest the use of Lindane. Interestingly the diagnostic ratio of β/(α + γ) HCH for more than 40% sites of BR and only one site of PR (PR-08, Moothakunnamb Harbor), was > 1 reflecting historical use of technical HCH formulation. Hence in Kerala we can see the combined impact of historical usage of technical HCH and ongoing usage of Lindane (Fig. 2). This observation is in line with urban and suburban transects of River Hooghly, India (Khuman and Chakraborty, 2019). However the average levels of γ-HCH were lower than River Hooghly and River Brahmaputra in India (Chakraborty et al., 2016a) and Shaying, (Bai et al., 2018) rivers in China (Bai et al., 2018) with the highest level of endosulfan sulfate (BR-02, 4.13 ng/L) in this study. Endosulfan sulfate was the only metabolite detected in specific networks, elevated concentrations of sum of β-endosulfan and the metabolite, endosulfan sulfate (ΣENDO) was observed in PR. The highest ΣENDO concentration was observed at Eloor (PR-07, 7.21 ng/L) in the midstream point of PR, with the dominance of β-endosulfan. It is to be noted that an insecticide industry is situated along the banks of PR in the Eloor industrial area and is considered as one of the world’s top toxic hotspots (Suchitra, 2020). Hence higher detection frequency of this highly persistent endosulfan isomer (β-endosulfan) can be possibly related to the industrial wastes along PR. Unlike PR, only a single sampling point at Thavanoor in the upstream section of BR was found with the highest level of endosulfan sulfate (BR-02, 4.13 ng/L) in this study. Endosulfan sulfate was the only metabolite detected in specific sites of CR. Owing to the low microbial degradation in the aqueous phase, endosulfan sulfate has higher persistence with a half-life of 187 days when compared with the parent isomers (Weber et al., 2010). Therefore, elevated levels of endosulfan sulfate in certain pockets of south India might be influenced by surface runoff from agricultural soil having a long history of endosulfan usage (Meire et al., 2016).

Mean concentration of ΣENDO in the rivers of south India was lower than levels reported from Gomti (Malik et al., 2009), Ganga (Khumand Chakraborty, 2019) and Ravi (Baqar et al., 2018) in Pakistan, River Karun in Iran (Behfar et al., 2013).

### 3.1.1.3. Endosulfan

We could not see any traces of α-endosulfan in south Indian rivers thereby indicating strict ban on technical formulation and absence of fresh input. Dominance of β-endosulfan in rivers of Kerala can be reasoned with extensive use in the recent past (Khuman et al., 2020) and a higher tendency of this isomer to undergo atmospheric deposition (Chakraborty et al., 2016a). Among the four river networks, elevated concentrations of sum of β-endosulfan and the metabolite, endosulfan sulfate (ΣENDO) was observed in PR. The highest ΣENDO concentration was observed at Eloor (PR-07, 7.21 ng/L) in the midstream point of PR, with the dominance of β-endosulfan. It is to be noted that an insecticide industry is situated along the banks of PR in the Eloor industrial area and is considered as one of the world’s top toxic hotspots (Suchitra, 2020). Hence higher detection frequency of this highly persistent endosulfan isomer (β-endosulfan) can be possibly related to the industrial wastes along PR. Unlike PR, only a single sampling point at Thavanoor in the upstream section of BR was found with the highest level of endosulfan sulfate (BR-02, 4.13 ng/L) in this study. Endosulfan sulfate was the only metabolite detected in specific sites of CR. Owing to the low microbial degradation in the aqueous phase, endosulfan sulfate has higher persistence with a half-life of 187 days when compared with the parent isomers (Weber et al., 2010). Therefore, elevated levels of endosulfan sulfate in certain pockets of south India might be influenced by surface runoff from agricultural soil having a long history of endosulfan usage (Meire et al., 2016).
and Chakraborty, 2019), Tapi (Hashmi et al., 2020), Tamirparani (Ariseker et al., 2021) rivers in India and River Karun in Iran (Behfar et al., 2013), Han, Geum, Yeoungsan, and Nakdong rivers in South Korea (Kim et al., 2020), and River Mudan in China (Wang et al., 2018). The average concentration of $\beta$-endosulfan in the south Indian rivers was lower than that of other rivers such as Brahmaputra and Hooghly (Chakraborty et al., 2016a; Khuman and Chakraborty, 2019) but comparable with Gomti in India (Malik et al., 2009) and Maozhou, Guanlan, Buji, Longgan and Pingshan rivers of China (Qiu et al., 2021). The levels were lower than levels from Chenab (Eqani et al., 2012) and Ravi (Baqar et al., 2018) rivers in Pakistan and Pampanga River, Philippines (Navarro et al., 2018) but were higher than levels observed in Quequén Grande River, Argentina (Silva-Barni et al., 2019) (Table S5).

3.1.1.4. Other pesticidal POPs. Selected sites in BR and PR were evidenced with trace levels of methoxychlor, cis, and trans-chlordane. The highest concentration of sum of endrin and its metabolites was observed at PR-9 (Munambam, 11.7 ng/L) with a dominance of the metabolite, endrin ketone thereby reflecting surface run-off from historical use in the agricultural fields. The average concentration of endrin ketone for the four rivers in south India was lower than Densu River, Ghana (Kuranchie-Mensah et al., 2012) and River Niger, Nigeria (Unyimadu et al., 2018). Heptachlor was not reported in any of the samples but its metabolite, heptachlor epoxide was detected at more than 50 % of the sites from BR and PR. Higher levels of this metabolite can be due to the prolonged heptachlor use in Kerala as a broad-spectrum insecticide (Gopalan and Chenicherry, 2018), coupled with the higher stability and aqueous solubility (Park and Bruce, 1968). The average concentration of heptachlor epoxide in BR and PR were comparable with levels reported from River Shaying, China (Bai et al., 2018) but was lower than that of lower stretch of Ganga in India (Khuman and Chakraborty, 2019). In surface water samples from Chennai, we found trace levels of dieldrin and heptachlor epoxide before the pandemic. However after the outbreak of the COVID-19 pandemic, elevated levels of aldrin, dieldrin, endrin ketone and heptachlor were observed in AR and CR.

3.1.2. Legacy industrial POPs

3.1.2.1. PCBs. Before the outbreak of the COVID-19 pandemic, the
average concentration (in ng/L) of sum of 25 congeners (Σ25PCBs) was highest in AR (33.27) followed by BR (21.84), CR (19.63) and PR (13.14). In the rivers of south India, the homologue profile was dominated by tetra congeners and PCB-44 was the dominant congener (BR=54 %, PR= 49 %, CR =64 %, AR =92 %) (Fig. 3, S1 and S2). PCB-44 is formed as an intermediate during the dechlorination of PCBs occurring in sewers, thereby making direct discharge of wastewater, and surface run-off as prime sources in the aqueous environment (Rodenburg et al., 2011). Low chlorinated PCBs (di to tetra chlorobiphenyls) in surface water might be associated with their relatively higher water solubility over the highly chlorinated PCBs (Rissato et al., 2006). A similar trend of dominance of lower chlorinated PCB homologues was observed in surface river water of Yamuna (Kumar et al., 2012), Hooghly and Brahmaputra (Chakraborty et al., 2016a) rivers in India, Ravi (Baqar et al., 2017) and Chenab (Eqani et al., 2012) rivers in Pakistan. In general, the PCB levels in rivers of southern India were lower than levels reported in the recent past from other Indian rivers viz., Hooghly (Σ19PCBs: 39–161 ng/L), Brahmaputra (Σ19PCBs: 57–233 ng/L) (Chakraborty et al., 2016a), and Yamuna (Σ27 PCBs: 2–779 ng/L) (Kumar et al., 2012) and lower than the coast of Bangladesh, (Σ209 PCBs: 32.17–199.4 ng/L) (Habibullah-Al-Mamun et al., 2019). It is to be noted that the current range of PCBs in surface water of south India has been found to be much higher than levels reported nearly a decade and a half back from the coastline of Chennai city along the Bay of Bengal, India (PCBs: 1.934–4.458 ng/L) (Rajendran et al., 2005). However, the PCB levels were in line with the observation from River Chenab, Pakistan (Σ24 PCBs: 7.7–110 ng/L) (Eqani et al., 2012) but several times higher than levels observed from the Himalayan Riverine Network (Σ33PCBs: 0.031–0.175 ng/L) (Ullah et al., 2020) and Indus River, Pakistan (Σ21 PCBs:0.003–0.2 ng/L) (Sohail et al., 2022), Yangtze River Delta (Σ38 PCBs:1.23–16.6 ng/L) (Zhang et al., 2011), Yellow River (average Σ33 PCB:0.2 ng/L) (Chen et al., 2021) and Songhua River (Σ43 PCBs:0.26–9.7 ng/L) (You et al., 2011) in China and Hudson River Estuary, USA (average Σ90 PCB:1.1 ng/L) (Yan et al., 2008).

The PCB dataset was subjected to multivariate PCA to identify specific sources associated with PCBs in surface water. The three principal components segregated accounted for 22 %, 18 % and 12 %, respectively of the total variance (Figure S3). PC–1 was loaded with 6 major mono-ortho dl-PCBs including PCB-123, PCB-105, PCB-118, PCB-157, PCB-156, PCB-189. Mean concentration of Σ12dl-PCBs in south Indian rivers are in line with the levels observed in River Hooghly and River Brahmaputra (Chakraborty et al., 2016a) but lower than River Yamuna (Kumar et al., 2012) in India and higher than Himalayan Riverine Network (Ullah et al., 2020). More than 90 % of dl-PCB loading in rivers from Kerala stemmed from PCB-105, – 118, – 157, – 156, – 189. Mean concentration of Σ12dl-PCBs in south Indian rivers are in line with the levels observed in River Hooghly and River Brahmaputra (Chakraborty et al., 2016a) but lower than River Yamuna (Kumar et al., 2012) in India and higher than Himalayan Riverine Network (Ullah et al., 2020). More than 90 % of dl-PCB loading in rivers from Kerala stemmed from PCB-105, – 118, – 123. The highest concentration of Σ12dl-PCBs was seen at Kanjoor and Aluva in PR at a close proximity (<2 kms) from Brahmapuram dumpyard (PR-4, 2.5 ng/L, and PR-5, 2.8 ng/L). In India, 70–80 % of waste ends up in smaller roadside street dumps or larger municipal solid waste dumpyards, wherein they are destined for open burning. Such dl-PCBs are usually associated with incomplete combustion processes including open burning of municipal solid waste in Indian cities (Chakraborty et al., 2021a, 2018). Street burning of waste in

![Fig. 3. Error plots (95 % confidence interval) showing the congener profiles for indicator and dioxin-like polychlorinated biphenyls (dl-PCBs) in Bharathappuzha and Periyar rivers of Kerala and Adyar and Cooum rivers of Tamil Nadu.](image-url)
Kerala was previously established to be a prominent emission source for these dl-PCB congeners both in the atmosphere and the burnt residues (Ajay et al., 2022; Sharma and Jain, 2019). It is to be noted that the most toxic PCB congener, PCB-126 was seen only in two sites (BC-1: 0.03 ng/L and PR-02: 0.3 ng/L) before the pandemic. Being located close to open dumps, PCB-126 in these sites might have resulted from the open burning of dumped plastic waste including e-waste in the waste stream (Chakraborty et al., 2018). In Chennai, more than 85 % of these dl-PCBs were found in surface water before the pandemic with > 50 % contribution from CR alone specifically from previously identified informal e-waste recycling areas of northern Chennai (Chakraborty et al., 2018). Informal e-waste recycling workshops were evidenced with fingerprints of such PCB congeners (Chakraborty et al., 2018). The informal e-waste recycling workshops were evidenced with fingerprints of such PCB congeners (Chakraborty et al., 2018). The informal e-waste recycling workshops were evidenced with fingerprints of such PCB congeners (Chakraborty et al., 2018). The diagnostic ratio of $p,p'$-DDT/1,1,1-Trichloroethane was > 1 at specific sites (ARc-02, ARc-03 and CRc-02). Apart from high detection frequencies of $p,p'$-DDT, a strong significant correlation ($R^2 = 0.829$, $p < 0.01$) indicating similarity in source profile. These two sites together contributed to > 60 % of PCB-18 in surface water samples from south India. Effluent discharge is a potential source of PCB-18 in water (Pham et al., 1999). Heavily polluted stretch of BR was evidenced with the highest concentration of these congeners specifically at Thavanoor (site BR-02, 77 ng/L), receiving a mixture of domestic and industrial wastewater (KWA, 2021). BR-09 is situated at the intersection with the Tirur Ponnani River which is known to receive a cumulative biochemical oxygen demand load of > 100 kg/day from open drains (KSRRC, 2019). Incidentally, BR-09 is also situated at close proximity to known open drains and receives heavy load of domestic wastewater (Suresh et al., 2020). PCB-138 and -180 shared similar detection frequencies with PCB-18 but with lower concentrations and related to wastewater discharge of industrial and domestic outfalls in several studies (Katsyvannis and Samara, 2004; Sakan et al., 2017; Tatarus et al., 2019). We found these indicator PCB congeners after the outbreak of pandemic in specific sites of CR (CRc-04, CRc-03) located adjacent to direct wastewater discharge outfalls. Influx of domestic sewage (80 × 10^3 liters/day) and industrial effluents (0.4 × 10^3 liters/day) from over 158 direct discharge outfalls might have impacted the load of these indicator PCB congeners in CR (Gowri et al., 2020). Hence congeners segregated in PC-2 can be attributed to wastewater intrusion via wastewater discharges or open drains. Among the 3 major PCB congeners (PCB-52, -28, -167) loaded in PC-3, PCB-28, and -52 contributed to one-third of the total PCB concentration in this study. The detection frequency of these congeners varied between 25 % and 43 % and PCB-52 alone contributed to > 80 % of PCBs segregated in PC-3. Furthermore, the average loading of the sum of these three congeners increased by 20-fold in rivers of Chennai after the outbreak of the pandemic. Open burning of domestic waste has been identified as a potential source of PCB-52 in the atmosphere (Chakraborty et al., 2021a) and soil of Indian cities (Chakraborty et al., 2018). It is noteworthy, that the highest concentration of 250 PCBs with dominance of PCB-52 followed by PCB-28 was evident after the outbreak of COVID-19 pandemic at a site in the upstream of the Buckingham canal (BC-c1, 166.68 ng/L), located within a 5 km radius of Kodungaiyur dumpsite. Dominance of atmospheric PCB-52 in urban Chennai was reasoned with open burning of solid waste at Kodungaiyur dumpsite (Priti Viraj and Chakraborty, 2020). Hence, surface run-offs or atmospheric deposition from the open burning of solid waste can be attributed as the major source of these tri and tetra PCB congeners in this study.

3.2. Variations in legacy and new POPs before and after the outbreak of pandemic

During pre-pandemic time, $p,p'$-DDT was not detected in the rivers of Chennai city. After the outbreak of the COVID-19 pandemic, ~75 % sites in AR and CR were detected with $p,p'$-DDT residues (Fig. 2, Table S4). The diagnostic ratio of $p,p'$-DDT/$p,p'$-DDE was > 1 at specific sites (ARc-02, ARc-03 and CRc-02). Apart from high detection frequencies of $p,p'$-DDT, a strong significant correlation ($R^2 = 0.829$, $p < 0.01$) indicating similarity in source profile. Elevated levels of PCB-52 in particular, has been evidenced as a fingerprint for open municipal solid waste burning in India (Chakraborty et al., 2018, 2016c). Atmospheric emission of this marker indicator PCB from an open municipal dumpsite was evident in urban Chennai (Priti Viraj and Chakraborty, 2020). Waste plastic materials dumped in the solid waste stream are interlinked with release of such industrial POPs in India (Chakraborty et al., 2022). Due to the rampant use of various personal protective equipment (PPE) made of plastics such as facial masks, gloves etc., a 17 % increase in the biomedical waste was reported from India (Robin et al., 2021). Tamil Nadu was one among the top five states in India producing the highest amount of plastic waste during the first wave of pandemic between June 2020 to November 2020 (Mallick et al., 2021). Burning of plastic waste might lead to an emission factor of up to 12, 446 ng/kg of PCBs (Wu et al., 2021). Hence such rise in the level of indicator PCBs during the pandemic especially after the lockdown phase can be attributed to open burning of dumped waste containing additional waste plastic from PPE. On the contrary, dl-PCB levels drastically decreased in the rivers of Chennai city after the outbreak of the pandemic and the levels in CR decreased by 4-fold. The bank of CR houses the core informal e-waste recycling workshops in northern part of Chennai with strong evidence of release of dl-PCBs in soil (Chakraborty et al., 2018), dust (Chakraborty et al., 2016b) and air (Chakraborty et al., 2021a). A significant decline ($p < 0.05$) in the concentration of specific dl-PCB congeners (PCB-105, -118, and -123) was seen in the e-waste sites during the COVID-19 pandemic when compared with levels before the outbreak of the pandemic. Hence we suggest that such decline in dl-PCBs is an indication of the temporary closure of informal e-waste recycling workshops and industrial sectors due to the intense lockdown in Chennai city. Furthermore, unlike past few years, Chennai experienced a greater impact of the northeast monsoon in 2020 (57.9 mm rainfall) (Skytemweather, 2021). Hence, the sudden spike of selected POPs after the outbreak of the
COVID-19 pandemic can be associated with atmospheric deposition and surface run-off from the potential fresh source regions in Chennai.

3.3. Flux estimation and risk assessment

Even though the flowrate of PR and BR are comparable, due to the higher POPs concentrations, BR exhibited higher fluxes (in tons/year) for HCH (0.03), DDT (0.006), and endosulfan (0.002) over HCH (0.01), DDT (0.009), and endosulfan (0.01) in PR. Similarly, the flux estimated for total PCBs was more than 2-fold higher in BR (0.15 tons/year) over PR (0.06 tons/year) with over 50% contribution by PCB-44 alone in both BR and PR. The fluxes estimated were lower than those reported from River Chenab in Pakistan (Eqani et al., 2012).

3.3.1. Eco-toxicological risk assessment due to OCPs

Eco-toxicological risks due to DDT, endosulfan, and HCH for different group of organisms including phytoplanktons, zooplanktons, molluscs, insects, and fishes are illustrated in Fig. 4. Phytoplanktons have been identified as an important pathway for OCPs bio-magnification towards higher trophic level organisms (Qiu et al., 2017). The average hazard quotient estimated revealed a major risk towards lower trophic level organisms like phytoplankton (diatoms) and zooplankton (scud) due to HCH and in > 30% sites of BR and PR due to DDT and endosulfan in daggerblade grass shrimp. In both BR and PR > 50% sites showed high risk due to DDT for insects' group specifically for mosquitoes. In PR, one third of the sites are having HQ values > 1 with significant impact of endosulfan on fishes including edible fish species such as Catla and Bluegill. Such impact of endosulfan on edible fish species in Kerala is consistent with the observations from the lower stretch of Ganga (Chakraborty et al., 2016a; Khuman and Chakraborty, 2019). In CR, high risk due to HCH was estimated for phytoplanktons, zooplanktons and insects like mosquitoes with HQ values > 1 in nearly 75% sites. After the outbreak of the pandemic, for most of the sites in the rivers of Chennai (CR and AR), DDT posed high risk for mosquitoes, thereby indicating the DDT usage for vector control.

3.3.2. Toxic equivalents (TEQs)

The range of TEQs for dl-PCBs in the rivers of south India are given in Table S7. Highest TEQ in pgTEQ/L was seen in PR (ND-30.396) followed by BR (0.0037–0.077), CR (ND-0.073) and AR (0.017–0.048) before the outbreak of the COVID-19 pandemic (Red dotted line indicates hazard limit given by USEPA).

Fig. 4. Error plots (95% confidence interval) showing eco-toxicological risk estimated from surface water concentrations of sum of isomers and metabolites of DDT, endosulfan and HCH isomers in Bharathappuzha and Periyar rivers of Kerala and Adyar and Cooum rivers before and after the outbreak of the COVID-19 pandemic (Red dotted line indicates hazard limit given by USEPA).
4. Conclusion

Dominance of \( p,p'\)-DDE, \( \beta\)-HCH and \( \beta\)-endosulfan in the rivers of south India reflected extensive use of these POPs in the past. Atmospheric deposition and surface run-off from the open burning of dumped waste and discharge of wastewater were the potential sources for tri and tetra PCBs in rivers of south India. Even though a distinct decrease in OCP levels was evidenced during the pandemic, the sudden spike in the pandemic suggested the frequent practice of open burning of dumped solid waste including profound increase of waste plastic from PPE usage in Chennai city. Impact of lockdown imposed after the outbreak of COVID-19 pandemic led to the closure of informal e-waste recycling sectors in Chennai leading to a decrease in the levels of dl-PCBs.

Environmental Implication

After the outbreak of the COVID-19 pandemic, due to lockdown there was a worldwide improvement in the water quality index in various water bodies. However, there is a need to understand the variations in levels and sources for persistent organic pollutants (POPs) before and after the outbreak of the COVID-19 pandemic. Hence, we took the first attempt to investigate chlorinated legacy and new POPs in four major rivers from Kerala and Tamil Nadu in southern India entering into the Arabian Sea and the Bay of Bengal, respectively.

CRediT authorship contribution statement

Ronnie Rex K: Formal analysis, Data curation, Writing – original draft. Paromita Chakraborty: Conceptualization, Methodology, Software, Supervision, Writing – review & editing, Fund acquisition.

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.

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Appendix A. Supporting information

Supplementary data associated with this article can be found in the online version at doi:10.1016/j.jhazmat.2022.129262.

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