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Organic contaminants in Ganga basin: from the Green Revolution to the emerging concerns of modern India

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PII: S2589-0042(21)00090-0

DOI: <https://doi.org/10.1016/j.isci.2021.102122>

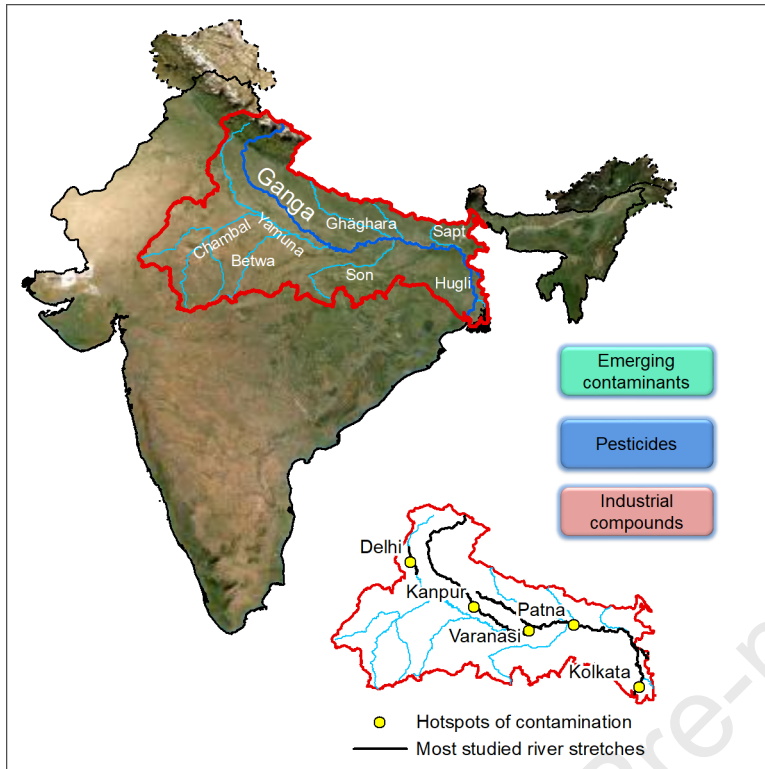
Reference: ISCI 102122

To appear in: *ISCIENCE*

Please cite this article as: Ghirardelli, A., Tarolli, P., Rajasekaran, M.K., Mudbhatkal, A., Macklin, M.G., Masin, R., Organic contaminants in Ganga basin: from the Green Revolution to the emerging concerns of modern India, *ISCIENCE* (2021), doi: <https://doi.org/10.1016/j.isci.2021.102122>.

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1 **Organic contaminants in Ganga basin: from the Green Revolution to the** 2 **emerging concerns of modern India**

3 4 **Commissioned Review Article**

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18 19 **Summary**

20
21 The Ganga basin includes some of the most densely populated areas in the world, in a region
22 characterised by extremely high demographic and economic growth rates. Although anthropogenic
23 pressure in this area is increasing, the pollution status of the Ganga is still poorly studied and
24 understood. In the light of this, we have carried out a systematic literature review of the sources,
25 levels and spatiotemporal distribution of organic pollutants in surface water and sediment of the
26 Ganga basin, including for the first time emerging contaminants (ECs). We have identified 61
27 publications over the past thirty years, with data on a total of 271 organic compounds, including
28 pesticides, industrial chemicals and by-products, artificial sweeteners, pharmaceuticals and personal
29 care products (PPCPs).

30 The most studied organic contaminants are pesticides, whereas knowledge of industrial compounds
31 and PPCPs, among which some of the major ECs, is highly fragmentary. Most studies focus on the
32 main channel of the Ganga, the Yamuna, the Gomti and the deltaic region, while most of the
33 Ganga's major tributaries, and the entire southern part of the catchment, have not been investigated.
34 Hotspots of contamination coincide with major urban agglomerations, including Delhi, Kolkata,
35 Kanpur, Varanasi and Patna. Pesticides levels have decreased at most of the sites over recent
36 decades, while potentially harmful concentrations of polychlorinated biphenyls (PCBs), organotin
37 compounds (OTCs) and some PPCPs have been detected in the last ten years. Considering the
38 limited geographical coverage of sampling and number of analysed compounds, this review
39 highlights the need for a more careful selection of locations, compounds and environmental
40 matrices, prioritizing PPCPs and catchment-scale, source-to-sink studies.

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41 1. Introduction

42

43 In recent decades, pollution of water bodies has become a matter of growing concern in the low and
44 middle income countries. Rapid industrialization and population growth have increased the release
45 of industrial and domestic effluents to surface water, jeopardizing aquatic ecosystems and
46 compromising water quality (Paul, 2017).

47 The Ganga basin, one of the most densely populated areas in the world with exceptionally high
48 population and economic growth rates (Census Data, 2011), is typical in this respect where the
49 widespread contamination of water bodies has become a growing concern. Sediment and water
50 carried by the Ganga and its tributaries represent a crucial resource for agriculture and many other
51 economic activities, directly or indirectly supporting the livelihood of over 400 million people
52 (Kumar, 2017).

53 Despite growing anthropogenic pressure in the catchment and severe water quality deterioration
54 (Dwivedi et al., 2018), the pollution status of the Ganga is still poorly studied. Recent reviews have
55 been either general summaries of pollution in the Ganga (Agarwal, 2015; Dwivedi et al., 2018),
56 only reporting the main sources of contamination and not analysing concentration trends, or broader
57 studies about the Indian context that do not consider the river basin as an independent hydrological
58 unit (Agarwal et al., 2015; Balakrishna et al., 2017; Chakraborty et al., 2014; Mathew and
59 Kanmani, 2020; Mohapatra et al., 1995; Philip et al., 2018).

60 The main sources of contamination in the Ganga and its tributaries are sewage, industrial effluent,
61 agricultural runoff and religious activities (Dwivedi et al., 2018). Several researchers have reviewed
62 the status of heavy metal residues in water and sediment (Paul, 2017; Singh Sankhla et al. 2018),
63 while the total organic carbon and the presence of coliforms are regularly monitored by the Indian
64 authorities for public health reasons (CPCB, 2013). However, less attention has been given to most
65 classes of organic compounds, both synthesized intentionally and formed as by-products of human
66 activities. Previous reviews generally focused on specific categories of contaminants (Goel et al.,
67 2013; Sinha and Loganathan, 2015).

68 In the light of this, we review in this paper the environmental status of the Ganga and its tributaries
69 in India, with particular reference to the spatiotemporal distribution of organic contaminants at a
70 basin scale. In addition to pesticides and common industrial compounds, this study includes a
71 specific focus on emerging contaminants (ECs) such as antibiotics, nonsteroidal anti-inflammatory
72 drugs (NSAIDs) and artificial sweeteners (ASWs), which to our knowledge have never been
73 systematically reviewed in the Ganga basin. We identify pollution hotspots as well as knowledge
74 gaps, in order to guide future research campaigns and management policies that need to be
75 implemented in the basin.

76

77 2. Study area

78

79 The Ganga basin is the largest catchment within the Indian sub-continent (NMCG, 2012), covering
80 an area of 1.086 million km² (CPCB, 2013), 79 per cent of which is located in India (Mirza,
81 2004). The Ganga originates from Gangotri glacier near Gomukh (Uttarakhand) where the
82 Bhagirathi river begins at an elevation of about 7010 m above mean sea level. The combined flow
83 formed at the confluence between the Bhagirathi and the Alaknanda, is known by the name Ganga

84 (*Sinha, 2004*). After flowing for over 2525 km through the plains of Uttarakhand, Uttar Pradesh,
 85 Bihar, Jharkhand, and West Bengal, the Ganga discharges into the Bay of Bengal. The Indian
 86 section of the Ganga delta conventionally begins after the Farakka barrage, close to the border
 87 between India and Bangladesh. Downstream of the barrage, the final reach of the main channel is
 88 known as Hugli (*Jain et al., 2007*). Along its course, the Ganga is joined by many tributaries, the
 89 longest of which is the Yamuna, which crosses the National Capital Territory (NCT, Delhi).

90 Water flow in the river system is highly seasonal due to the Indian Summer Monsoon: about 84 per
 91 cent of the total rainfall occurs in the monsoon season, from June to September (*CWC and NRSC,*
 92 *2014*).

93 With its 450 million inhabitants, the Ganga basin is one of the most populous regions on Earth
 94 (*Kumar, 2017*). According to the *2011 Census Data*, the average population density in the Ganga
 95 basin is 520 persons per square kilometre, as compared to 312 for the rest of India. In the delta
 96 zone, the average population density rises to over 900 people per square kilometre. Since the mid-
 97 20th century, the population of the eleven Indian states comprising the Ganga basin has grown
 98 considerably from 170 million people in 1951 to 611 million people in 2011 (*Census Data, 2011*).
 99 In the 21st century, demographic growth has particularly affected urban areas, where population
 100 increased by 30 per cent between 2001 and 2011.

101 The Ganga basin is also the primary contributor to the agricultural economies of India, thanks to the
 102 availability of fertile soils across the region (*NMCG, 2011*). As a consequence, more than 65 per
 103 cent of the basin area is covered with agricultural land (*CWC and NRSC, 2014*). Besides agriculture,
 104 hundreds of industrial plants are situated in the basin, comprising thermal power plants, electric
 105 industries, textiles, wood and jute mills, sugar mills, distilleries, pulp and paper factories, synthetic
 106 rubber industries, dairies, coal washeries, pesticide factories, and tanneries (*Dwivedi et al., 2018*).
 107 The major industrial centres of the basin, with around 1000 production units are located in Uttar
 108 Pradesh. The biggest industrial cities are concentrated in the area from Kannauj to Varanasi: the
 109 leading economic activities in Kanpur, Allahabad and Varanasi are focused on tannery, engineering,
 110 carpets and locomotive sectors (*Dwivedi et al., 2018*).

111 Two of the world's largest industrial cities, Kolkata and Delhi, with 14.0 million and 16.35 million
 112 inhabitants, respectively, are located in the Ganga basin (*Census Data, 2011*).

113

114 **Figure 1.**

115

116 **3. Organisation of the database and selected bibliography**

117

118 Only articles whose study area fell within the watershed of the Ganga (as defined by India Water
 119 Resources Information System (*CWC and NRSC, 2014*), **figure 1**) were considered in this review.
 120 Primary data related to river sediment and surface water were selected, the latter comprising river,
 121 pond, artificial canal and reservoir water bodies. A total of 61 papers provided primary data on the
 122 occurrence of organic contaminants in surface water and river sediment. Out of these, 28
 123 publications assessed surface water quality, 21 sediment and 12 analysed both water and sediment.
 124 Besides the Ganga itself, most of the sampling areas are located along the Yamuna, the Gomti and
 125 the delta (Hugli reach) (**figure 2 A-C**), coinciding with big urban agglomerations such as Delhi,
 126 Kanpur, Allahabad, Varanasi, Patna and Kolkata. The time period of this review covers the last 33

127 years, from 1986 (when the earliest analysed paper was published) to 2019. Fifty out of 61 articles
128 were published after 2000, showing a growing interest in Indian environmental issues in the new
129 millennium.

130 A total of 261 individual organic compounds and 10 groups of compounds (detected as cumulative
131 concentrations) are reported and these are classified into three broad categories: emerging
132 contaminants (including pharmaceuticals, PCPs, caffeine, ASWs, parabens, phthalate plasticizers,
133 benzotriazoles, bisphenol A and PFAS), pesticides (including organochlorine pesticides (OCPs),
134 organophosphates (OPhs), pyrethroids, herbicides and fungicides) and industrial compounds
135 (including polychlorinated biphenyls (PCBs), polybrominated diphenyl ethers (PBDEs), organotin
136 compounds (OTCs) and polycyclic aromatic hydrocarbons (PAHs)).

137

138 **4. Emerging contaminants**

139

140 According to the definition provided by the United States Geological Survey (USGS), ECs are
141 “Any synthetic or naturally occurring chemical or any microorganism that is not commonly
142 monitored in the environment but has the potential to enter the environment and cause known or
143 suspected adverse ecological and/or human health effects” (USGS, 2016). Many substances used in
144 daily life, ranging from pharmaceuticals to detergents fall under this description (Philip et al. 2018;
145 Sharma and Kapoor 2014; Stuart et al. 2012).

146 Within pharmaceuticals, antibiotics are receiving increasing attention because of their ability to
147 induce the development of antibiotic resistance in pathogenic bacteria (Kümmerer, 2009). Besides
148 antibiotics, NSAIDs (e.g. diclofenac and ibuprofen) and other drugs such as acetaminophen
149 (paracetamol) and carbamazepine (an anti-epileptic compound), are emerging as possible threats to
150 aquatic ecosystems. Their effects on biota range from physiological to behavioural alterations
151 (Brodin et al., 2014; Klimaszuk and Rzymiski, 2018). In addition, NSAIDs are known for their
152 toxicity on avian species, first reported in scavenger birds of the Indian sub-continent (Cuthbert et
153 al., 2007; Naidoo et al., 2010). Also PCPs, employed as active substances or preservatives in
154 cosmetics, body care products, surfactants, detergents, insect repellents and sunscreen agents have
155 been widely studied in relation to their detrimental effects on aquatic biota (Champagne, 2009;
156 Stuart et al., 2012) and antimicrobial resistance (Scientific Committee on Emerging and Newly
157 Identified Health Risks, SCHENIR, 2009).

158 A major concern raised by the presence of pharmaceuticals and personal care products (PPCPs) in
159 aquatic environments is their ability to interfere with the endocrine system, altering its normal
160 functioning (Ebele et al. 2017). A primary example of such compounds, referred to as endocrine
161 disruptors (World Health Organization and United Nations Environmental programme, WHO and
162 UNEP, 2013), are steroid hormones (Irwin et al., 2001; Jobling et al., 1998; Länge et al., 2001;
163 Purdom et al., 1994; Tyler et al., 2005), whose presence in the aquatic environment can be related
164 both to natural excretion and to synthetic estrogens and progestogens used in animal husbandry
165 (Kuster et al., 2004) and for medical purposes (Monteiro and Boxall, 2010).

166 Besides PPCPs, other compounds have been widely reported to exhibit endocrine-disrupting
167 properties, such as bisphenol A (Eladak et al., 2015; Rezg et al., 2014; Rochester, 2013;
168 Vandenberg et al., 2012), an essential component of epoxy resins (Eladak et al., 2015), and
169 phthalates, mainly employed as plasticizers (Petrović et al., 2001).

170 ASWs are one of the most recently recognized classes of high-priority ECs among non-PPCPs, as
171 they are frequently detected in different environmental matrices (Luo *et al.*, 2019). Saccharine,
172 cyclamate, acesulfame K and sucralose are the most studied compounds. Although their ecotoxicity
173 is still poorly understood (Luo *et al.*, 2019), they are viewed as ideal indicators of domestic
174 wastewater contamination in surface and groundwater (Tran *et al.*, 2014).

175 In this study, compounds have been included in the class of ECs based on literature definitions, but
176 also on the basis that they are not yet included in routine monitoring campaigns in India, and that
177 first recordings of these chemicals in the Ganga basin are very recent in comparison with pesticides
178 and industrial compounds (ICs, **tables 1-6** and **S4-9**).

179
180 **Figure 2.**

181 182 **4.1 Location of sample points**

183
184 Only 13 papers evaluate ECs, but the majority of them analysed simultaneously different sub-
185 categories, including PPCPs, with the prevalence of biocides, antibiotics and NSAIDs.

186 The sample points for PPCPs are all concentrated in the main channel of the river system (i.e.,
187 especially around the cities of Kanpur, Allahabad, Varanasi and Patna, located in the middle reach
188 of the Ganga basin, and in the Hugli and deltaic region (**figure 2 A**, **table 1-2**). Besides the main
189 channel, papers mainly focused on the NCT in the Yamuna sub-catchment. In addition, Sharma *et al.*
190 (2019) also monitored the rivers Alaknanda and Bhagirathi in the Himalayan reach. The
191 remaining publications focused on the cities along the Gomti river (Nag *et al.*, 2018), in Ujjain
192 (Madhya Pradesh) (Diwan *et al.*, 2018) and Udaipur (Rajasthan) (Williams *et al.*, 2019).

193 However, the distribution pattern for surface water sampling differs considerably from sediment
194 sampling areas: papers reporting on water pollution were focused on big urban agglomerates such
195 as Delhi (Mutyar *et al.*, 2018; Mutiyar and Mittal, 2012, 2014a), Kanpur, Allahabad, Varanasi,
196 Patna (Sharma *et al.*, 2019) and Lucknow (Nag *et al.*, 2018). In the case of sediment, addressed
197 only by Nag *et al.* (2018), Diwan *et al.* (2018), and Chakraborty *et al.* (2019), sample points were
198 located along the Gomti river, in the city of Ujjain, and the Hugli area.

199 Some of the PCPs have been evaluated by only one paper, and in restricted reaches of the basin:
200 synthetic detergents (anionic surfactants) in sediment samples collected in Kolkata district (Ghose
201 *et al.*, 2009), musk fragrances and parabens in sediment along the Hugli (Chakraborty *et al.*, 2019).

202 With regard to non-PPCP compounds, the presence of ASWs has been reported only by Sharma *et al.*
203 (2019) in the main channel and the Himalayan rivers, whereas benzotriazole and
204 methylbenzotriazole have been reported by Williams *et al.* (2019) near Udaipur.

205 The distribution of bis (2- ethylhexyl) adipate plasticizers has been studied by Chakraborty *et al.*
206 (2019) along the Hugli, while phthalates have been assessed in sediment both in the Gomti
207 (Srivastava *et al.*, 2010) and the Hugli (Chakraborty *et al.*, 2019). Bisphenol A has been studied
208 both by Chakraborty *et al.* (2019) in the Hugli and by Williams *et al.* (2019) in surface water near
209 Udaipur. Levels of PFAS has been evaluated by three papers: Yeung *et al.* (2009) and Sharma *et al.*
210 (2016a) focused on water samples from cities and towns located along the main channel, the
211 Alaknanda, the Bhagirathi and the confluence between Ganga and Yamuna; Corsolini *et al.* (2012)
212 studied sediment contamination in the Hugli river and adjacent Sundarban wetlands.

213

214 4.2 Occurrence of ECs

215

216 The maximum concentrations of PPCPs in water exhibited a wide range from less than one to
217 thousands of ng/L, while sediment concentrations varied between less than one and hundreds of
218 µg/kg.

219 With regard to pharmaceuticals, the compound with the highest water concentration was the
220 antibiotic ampicillin (maximum recorded value, MRV: 27,100 ng/L, Delhi (*Mutiyar and Mittal,*
221 *2014b*)). For the NSAIDs ibuprofen had the highest values (MRV: 2302 ng/L, Delhi (*Mutiyar et al.,*
222 *2018*)), and in the hormone group, the highest concentration reported was for androsterone (MRV:
223 1557 ng/L, Udaipur (*Williams et al., 2019*)). The PCP with the highest concentration in water was
224 triclosan (MRV: 9650 ng/L, Gomti river (*Nag et al., 2018*)). The only reported value for surfactants
225 was a cumulative concentration comprising all the methylene-blue-active substances: not
226 surprisingly, it was higher than any single compound among PPCPs (MRV: 0.425 mg/L (425,000
227 ng/L), Kolkata (*Ghose et al., 2009*)).

228 Only six antibiotics, three NSAIDs, carbamazepine, musk ketone, four parabens and triclosan have
229 been assessed in river sediment (**table 2** and **S5**). The highest recorded concentration for
230 pharmaceuticals was 519 µg/kg dry weight (d.w.) (carbamazepine), followed by the NSAID
231 ibuprofen (MRV: 340 µg/kg d.w., Hugli river (*Chakraborty et al., 2019*)); the MRV for antibiotics
232 was 9.74 µg/kg d.w. (Ujjain, (*Diwan et al., 2018*)). The highest PCP value was recorded for methyl
233 paraben (MRV: 423 µg/kg d.w., (*Chakraborty et al., 2019*)), whereas triclosan and musk ketone
234 showed much lower concentrations (MRVs: 84 and 26 µg/kg d.w. respectively, (*Chakraborty et al.,*
235 *2019*)).

236 The highest concentration of non-PPCPs in water was found for caffeine (maximum recorded value,
237 MRV: 37,476 ng/L, Udaipur (*Williams et al., 2019*)), and the highest sediment concentration was
238 detected for di-(2-ethylhexyl) phthalate (MRV: 400 µg/kg d.w.), in the Hugli river (*Chakraborty et*
239 *al., 2019*)). ASW maximum water concentrations were extremely low compared to other PPCPs
240 sub-categories: the highest recorded value was 85.43 ng/L, found in Patna for saccharine (*Sharma et*
241 *al., 2019*). In the case of PFAS, the highest water concentration was found for perfluorobutane
242 sulfonate (PFBS) (MRV: 10.19 ng/L, Gangasagar (*Sharma et al., 2016*)), whereas the MRV for
243 sediment was 14.09 µg/kg d.w. (PFOA, Sundarban wetland (*Corsolini et al., 2012*)).

244 With regard to the spatial distribution, PPCPs analysed by more than one paper, in different areas of
245 the basin, are characterised by a wide range of variability in river water. Concentrations in the main
246 channel and in headwater rivers are generally below 10 ng/L, and often close to their limit of
247 detection (usually 0.1-5 ng/L). Water concentrations tend to be considerably higher downstream of
248 Delhi and in Udaipur: this pattern is evident for compounds such as acetaminophen, carbamazepine,
249 ciprofloxacin, DEET, diclofenac (not detected in Delhi, but very high in Udaipur),
250 hydrochlorothiazide, ibuprofen, naproxen and sulfamethoxazole. The latter exhibited higher water
251 concentrations also in Ujjain, and triclosan along the Gomti. **Figure 3** shows the variations in levels
252 of carbamazepine and sulfamethoxazole in the Ganga, selected to be representative of ECs.

253 Although the main Ganga channel was characterised by low concentrations (often below 10 ng/L),
254 *Sharma et al. (2019)* recorded that the analysed PPCPs were generally higher in middle and lower
255 reaches compared to the Himalayan reach, most notably downstream of major cities such as
256 Kanpur, Varanasi, and Patna. This pattern, also evident in compounds such as carbamazepine,

257 hydrochlorothiazide, sulfamethoxazole and diethyltoluamide (DEET, **figure S1**), is likely to result
258 from local releases of sewage and industrial wastewater, which are the main sources of ECs in
259 water bodies. Pollution loads do not increase along the main channel (lower concentrations are
260 recorded between Farakka and Gangasagar), and this behaviour is likely to arise from natural
261 attenuation processes (Narain, 2014). This has been observed on a smaller scale both in the Gomti
262 (Nag et al., 2018) and the Kshipra (Diwan et al., 2018), where point sources predominate.

263 Similarly to what recorded by Sharma et al. (2019) for PPCPs, also Yeung et al. (2009) and Sharma
264 et al. (2016) reported an increase in water concentration of PFAS up to the middle reach. The
265 exception to this pattern is shown by caffeine, whose concentrations in the Himalayan reach of the
266 Ganga were comparable to those detected in the middle and lower reaches of the basin (hundreds of
267 ng/L). As a whole, its concentrations are generally higher than other ECs, due to the very large-
268 scale consumption of this compound, which is common in food and beverages (Mutiyar et al.,
269 2018).

270 As far as sediment concentrations are concerned, the few ECs for which more than one article was
271 found showed comparable concentrations both along the Gomti (Nag et al., 2018; Srivastava et al.,
272 2010) and the Hugli (Chakraborty et al., 2019): phthalates maximum concentrations were in the
273 order of 300 ng/L, while triclosan maximum values ranged between 50 and 80 ng/L.

274

275 **Figure 3.**

276

277 **5. Pesticides**

278

279 Pesticides represent the most studied class of organic contaminants in the Ganga basin. They are a
280 direct consequence of so called “Green Revolution”, which resulted the widespread adoption of new
281 technologies and pesticides in agriculture in the 1970s.

282 The use of pesticides in modern agriculture has led to worldwide nonpoint pollution in aqueous
283 environments, affecting water bodies used for drinking water (Schulz, 2004; Zhang and Zhang,
284 2011) and nontarget organisms (Barranger et al., 2014; Ma et al., 2006; Ogbeide et al., 2015;
285 Stachowski-Haberkorn et al., 2013). However, pesticides can also originate from urban
286 environments: in particular, household agents used for control of vector-borne diseases such as
287 malaria or Leishmaniasis (Sinha and Loganathan, 2015).

288 In India, whose pesticide consumption accounts for just 3.75 per cent of worldwide use, 80 per cent
289 is represented by insecticides, 15 per cent by herbicides and 2 per cent by fungicides (Agarwal et
290 al., 2015).

291 Although growing environmental and human health risks have led to worldwide bans of numerous
292 pesticides (UNEP, 2018), they remain a matter of concern due to their high persistence and
293 ubiquitous presence in ecosystems and the environment.

294 A prominent example of this is represented by OCPs, a class of insecticides and acaricides that
295 include 9 of the first 12 contaminants listed in the Stockholm Convention on Persistent Organic
296 Pollutants (UNEP, 2018). Like all Persistent Organic Pollutants (POPs), OCPs such as DDT and
297 lindane are characterised by high hydrophobicity, lipophilicity and persistence in the environment,
298 (Zitko, 2003) and tend to bioaccumulate in the fatty tissues of biota, especially at high trophic levels
299 (Ntow et al., 2008). However, the use of DDT is still permitted in some regions of the world,

300 including India, for applications against mosquitoes to control malaria. Similarly, lindane (γ -
301 hexachlorocyclohexane, HCH isomer) can be employed for the control of body parasites (head lice
302 and scabies) (UNEP, 2018).

303 OPhs are another class of insecticides and acaricides potentially harmful for a wide variety of non-
304 target species (Goel *et al.*, 2013) and responsible for frequent cases of human poisoning in India
305 (Jokanović, 2018).

306

307 **5.1 Location of sample points**

308

309 As a result of their wide use in the Ganga basin since the Green Revolution, pesticides are the most
310 studied class of organic contaminants, with 33 papers reviewed in this study (tables 3-4 and S6-7).
311 The most frequently reported pesticides were OCPs, which have been documented along the entire
312 course of the main channel and the upper reaches since the 1970s (Bakre *et al.*, 1990). Study areas
313 for pesticides are not homogeneously distributed in the Ganga basin; the majority are focused on the
314 northern-central section of the basin, along the main channel, around cities such as Kanpur, Unnao,
315 Allahabad, Varanasi and Patna, and in the Himalayan reach of the Ganga (Alaknanda, Bhagirathi
316 and different streams). The Gomti sub-catchment, Delhi and the surrounding districts of Uttar
317 Pradesh along the Yamuna, and the Hugli and Sundarban wetlands have also been investigated. To
318 the best of our knowledge, south-bank rivers other than the Yamuna have not been assessed for
319 pesticides (figure 2 B).

320 OCPs belonging to the DDT, endosulfan and HCH group (main isomers and related metabolites)
321 and cyclodiene insecticides (aldrin, dieldrin, endrin) have been extensively analysed in water and
322 sediment along the entire course of the main channel and the rivers of the Himalayan region
323 (Ahmad *et al.*, 1996; Mutiyar and Mittal, 2013; Nayak *et al.*, 1995; Raghuvanshi *et al.*, 2014;
324 Rehana *et al.*, 1996, 1995; Sankararamkrishnan *et al.*, 2005; Sarkar *et al.*, 2003; Semwal and
325 Akolkar, 2006; Senthilkumar *et al.*, 1999; Singh *et al.*, 2012). Studies on the Yamuna and its canals,
326 however, were focused on the area around Delhi (Agarwal *et al.*, 1986; Aleem and Malik, 2005;
327 Kumar *et al.*, 2011; B. Kumar *et al.*, 2012b; Nair *et al.*, 1991; Pandey *et al.*, 2011) and the
328 surrounding agricultural regions of Haryana, including Western Yamuna, Agra and Gurgaon canals
329 (Kaushik *et al.*, 2008). Only one paper has investigated the presence of the OCPs in the upper reach
330 of Yamuna (Semwal and Akolkar, 2006). Water and sediment of the Hugli and Sundarban wetlands
331 have been addressed by five papers ((Bhattacharya *et al.*, 2003; Ghose *et al.*, 2009; Guzzella *et al.*,
332 2005; Mondal *et al.*, 2018; Sarkar *et al.*, 2008), the latter also sampling pond water). In addition,
333 two studies assessed the presence of OCPs in water and sediment along the Gomti (Malik *et al.*,
334 2009; Trivedi *et al.*, 2016).

335 Bakre *et al.* (1990) investigated the presence of HCH, Heptachlor and Aldrin in the waters of
336 Mahala reservoir, near Jaipur; Dua *et al.* (1996) focused on the distribution of DDT and HCH in 22
337 ponds in the district of Shahjahanpur, Uttar Pradesh; Dua *et al.* (1998) detected DDT and HCH
338 compounds in Bhimtal, Sattal, Khurpatal, Naukuchiatal and Nainital lakes (Nainital Himalayan
339 region); Singh *et al.* (2007) addressed HCB and several compounds belonging to DDT, HCH,
340 endosulfan, heptachlor, chlordane and cyclodiene groups in streams, ponds and canals between
341 Kanpur and Lucknow; Bishnu *et al.* (2009) studied the presence of heptachlor, dicofol and
342 endosulfan in waterbodies adjacent to the tea gardens of Dooars and Hill regions, West Bengal;

343 *Singh Bhadouria et al. (2012)* focused on compounds belonging to DDT, HCH, endosulfan,
344 heptachlor, chlordane and cyclodiene group, in the wetlands outside and inside Keoladeo National
345 Park, Rajasthan; *Rao and Wani (2015)* investigated the presence of DDT, HCH, HCB, endosulfan,
346 heptachlor, and cyclodiene pesticides in Tighra reservoir, near Gwalior (Madhya Pradesh). In
347 addition, *Jit et al. (2011)*, assessed the presence of HCH isomers in the Sharda and Reetha rivers,
348 and in drains surrounding a lindane factory located in Lucknow district. The major contaminants in
349 terms of records and spatial abundance of sample points were aldrin among the cyclodiene group,
350 p,p'-DDT among the DDT group, γ -HCH among the HCH group, and both α - and β -endosulfan
351 among the endosulfan group.

352 OPhs have only been detected around villages and cities located along the main channel, in the
353 upper-middle reach, namely Kachla, Narora, Fatehgarh, Kannauj and Kanpur (*Rehana et al., 1996,*
354 *1995; Sankararamkrishnan et al., 2005*). In addition, the presence of OPhs has been investigated
355 in the Hugli river and the surrounding ponds by *Mondal et al. (2018)*, and in the Yamuna by *Aleem*
356 *and Malik (2005)* and *Kumar et al. (2011)*, who exclusively focused on NCT.

357 Herbicides, such as alachlor, atrazine and butachlor, have been studied in sediment, river and pond
358 water from four sites along the Hugli (*Mondal et al., 2018*), in drains discharging into the Yamuna
359 in Delhi (*Kumar et al., 2011*), and along the Gomti, in the area of Lucknow (*Trivedi et al., 2016*).
360 2,4-dichlorophenoxyacetic acid (2,4-D) has been detected along the main channel in the upper-
361 middle reach (*Rehana et al., 1996, 1995*) and NCT along the Yamuna (*Aleem and Malik, 2005*).

362 Both fungicides and pyrethroids have been detected only in the Hugli and in ponds of the deltaic
363 region (*Mondal et al., 2018*), as well as in streams, ponds and canals between Kanpur and Lucknow
364 (*Bishnu et al., 2009*).

365

366 **5.2 Occurrence of pesticides**

367

368 Pesticides showed the greatest variability both in water and sediment, with values ranging from less
369 than one ng/L to mg/L and from less than one to thousands of $\mu\text{g}/\text{kg}$ d.w. ($\mu\text{g}/\text{kg}$), respectively.

370 For OCPs, the compounds with the highest concentration were α -HCH for water (MRV: 0.29 mg/L
371 (290000 ng/L) at Lucknow (*Jit et al., 2011*)) and γ -HCH for sediment (MRV: 7540 $\mu\text{g}/\text{kg}$ d.w. at
372 Bharatpur (*Singh Bhadouria et al., 2012*)).

373 Among OPhs, the highest water concentration was found for malathion (MRV: 2610 ng/L, Kanpur
374 (*Sankararamkrishnan et al., 2005*)), whereas the MRV for sediment was 458.02 $\mu\text{g}/\text{kg}$ d.w.
375 (methyl parathion, Bhagalpur (*Singh et al., 2012*)).

376 The only available paper on pyrethroids (*Bishnu et al., 2009*) found concentrations below the
377 detection limit. Similarly, the only analysed fungicide (metalaxyl), was below the detection limit in
378 sediment and 83 ng/L in water (final reach of the Hugli (*Mondal et al., 2018*)).

379 Herbicides with the highest concentration were butachlor in water (MRV: 0.135 mg/L (135000
380 ng/L), Lucknow (*Trivedi et al., 2016*)) and pendimethalin in sediment (MRV: 53.19 $\mu\text{g}/\text{kg}$ d.w.,
381 Delhi (*Kumar et al., 2011*)).

382 In terms of spatial distribution, the Himalayan districts exhibited low concentrations of pesticides
383 compared to the main Ganga channel, and from samples taken from artificial canals (e.g. Western
384 Yamuna, Agra and Gurgaon canals). This is particularly the case for OCPs, such as DDT and HCH.

385 However, no upstream-downstream trend was detected in the Ganga and contamination levels
386 appear to be influenced by local pollution sources and attenuate quite rapidly downstream as
387 consequence of dilution or adsorption by river channel sediment (Narain, 2014).

388 High concentrations were found in the proximity of big urban agglomerates such as Delhi,
389 Allahabad, Varanasi and Lucknow. It has been reported that cities located along the Ganga and its
390 tributaries have contributed to OCP pollution mainly through the release of insecticides in
391 wastewater during vector-control campaigns (Trivedi et al., 2016).

392 However, the main Ganga channel is characterised by lower concentrations compared to canals,
393 ponds and lakes, as recorded by Singh et al. (2007) and Jit et al. (2011) in the plain between Kanpur
394 and Lucknow. This pattern, evident in water, is less noticeable in sediment, probably due to the
395 more limited availability of papers, which prevents any detailed spatial assessment. In addition,
396 spatial comparisons would require accurate knowledge of the organic carbon content of the matrix,
397 that can be highly variable in different locations. In fact, as POPs tend to be associated with
398 organic-rich particles, sediment concentration values are influenced by organic carbon content
399 (Binelli et al., 2007).

400 An overall decrease in pesticide concentration is evident in surface water, as previously reported by
401 (Dwivedi et al., 2018). The declining trend over the 40-year timeframe of the studies was evident
402 especially for persistent pesticides, whose bans and limitations have positively affected the
403 environmental status of the Ganga. However, no clear trend was shown for sediment. This might be
404 due to multiple reasons, including the fewer number of papers that have studied sediment pollution
405 and the different pollution dynamics in terms of mass load and flow rate in the two matrices.

406 Focusing on the two most studied and detected pesticides, DDT and HCH (isomers and
407 metabolites), these showed similar spatial and temporal concentration trends. This reflects the
408 comparable use and history of the two compounds in the region with both extensively used in
409 agriculture since the Green Revolution, but also employed for sanitation purposes after restrictions
410 introduced in the 1990s (Yadav et al., 2015). However, there is a decrease in concentrations of both
411 pesticides after 2010 (figure 4 and 5).

412

413 **Figure 4.**

414

415 **Figure 5.**

416

417 **6. Industrial compounds**

418

419 This category of environmental contaminants includes a variety of compounds synthesized or used
420 in chemical plants and other manufacturing processes, or released as industrial by-products. For
421 some of them, such as OTCs, pollution results from the disposal and breakdown of manufactured
422 products. Unlike ECs, these contaminants have been studied in the Ganga Basin since the 1990s, or
423 even the 1980s in the case of PAHs. These compounds are well-known for their detrimental health
424 and environmental effects and have already been regulated by international and Indian authorities.
425 PAHs and PCBs are now periodically monitored in India and have acceptable limits of
426 contamination defined by the Indian drinking water quality standards (Bureau of Indian Standards
427 (BIS, 2012)). Furthermore, in 2015 India complied with the International Convention on the Control

428 of Harmful Anti-fouling Systems on Ships (AFS) (*International Marine Organisation, IMO, 2018*).
429 Most of these chemicals are included in the class of POPs listed in the Stockholm Convention,
430 ratified by India in 2006 (*UNEP, 2018*). Among them are PBDEs, specifically tetra-, penta-, hexa-,
431 hepta- and decabromodiphenyl ether, belonging to the class of brominated flame retardants
432 (*Rahman et al., 2001*).

433
434 PCBs are officially recognized as POPs (*UNEP, 2018*). Employed in many industrial applications
435 (e.g. transformers, capacitors, paints and printing inks) (*Erickson and Kaley, 2011*), PCBs are
436 released into the environment through leaks or fires, and spills during the transport of products
437 containing them (*S. Kumar et al., 2012*).

438 Despite not being listed in the Stockholm Convention, many analogies can be found between PAHs
439 and POPs, as they share lipid solubility and persistence in the environment (*Abdel-Shafy and*
440 *Mansour, 2016*). PAHs may result from a series of combustion processes, including pyrolysis of
441 wood to produce charcoal and carbon black, power generation from fossil fuels, incineration of
442 waste, vehicular emissions (*Malik et al., 2011*). PAHs are well-known mutagens and teratogens and
443 human carcinogens (*Abdel-Shafy and Mansour, 2016*).

444 Another common link between different classes of ICs is their ability to act as endocrine disruptors.
445 One of the most studied examples of endocrine disruption in wildlife is imposex induced in
446 gastropods by OTCs (*Matthiessen, 2013; Sousa et al., 2014*). The most notorious of these chemicals
447 is tributyltin TBT, a biocide used in antifouling paints, which can negatively affect non-target
448 aquatic organisms (*Bangedphol et al., 2009; Garg et al., 2011; Ohji et al., 2007*). Also PCBs have
449 been reported as endocrine disruptors (*Sharma and Kapoor, 2014*).

450

451 **6.1 Location of sample points**

452

453 Among the analysed studies, 20 papers assessed ICs, with most (10 publications) reporting PAHs
454 (**tables 5-6 and S8-9**). As with ECs and pesticides, the geographical distribution of sample points
455 for ICs is uneven, with most studies focused on the main channel of the Ganga, in the cities of the
456 upper and middle reaches (e.g. Kanpur, Allahabad, Varanasi and Patna). Besides the Ganga, studies
457 were concentrated along the Gomti, in Delhi and the nearby districts of Haryana and Uttar Pradesh.
458 The largest concentration of sample points is located in the Hugli reach and the deltaic region
459 (**figure 2 C**), but there is no available data for south-bank rivers other than the Yamuna.

460 The distribution of study areas of PAHs in surface water considerably differs from sediment. The
461 former include a greater number of papers analysing the Gomti and the Himalayan rivers, whereas
462 in the case of sediment, more attention was given to the central area of the main channel and the
463 deltaic region, but with no data on the Himalayan reach of the Ganga. PAHs have mainly been
464 assessed in sediment from the Hugli river and from Sundarban (*Binelli et al., 2008; Guzzella et al.,*
465 *2005; Zuloaga et al., 2013*), in water and sediment along the Gomti, in the towns of Neemsar,
466 Bharatpur, Lucknow, Barabanki, Sultanpur and Jaunpur (*Malik et al., 2011, 2004; Tripathi et al.,*
467 *2009*), and in eight cities and towns located along the Alaknanda, the Bhagirathi, and the main
468 channel (Uttarkashi, Devprayag, Narora, Kachala, Fatehgarh, Kannauj, Kanpur, Varanasi, Patna,
469 Farakka), as well as in the delta island of Gangasagar, and the Gangotri glacier (*Ahmad et al., 1996;*
470 *Sharma et al., 2018*). Only one paper assessed the presence of PAHs in the Yamuna river,

471 specifically in sediment upstream and downstream of Dehli (*Agarwal et al., 2006*), while one paper
472 addressed the presence of PAHs in core samples taken in Nainital and Bhimtal Lakes, located in the
473 Himalayan region of the basin (*Choudhary and Routh, 2010*).

474

475 Spatial patterns of other IC categories in surface water and sediment are similar, although many
476 water studies were concentrated in Dehli and urban areas located along the main channel, whereas
477 most of the papers addressing sediment were focused on the Hugli river and its estuary. PBDEs
478 have only been investigated in sediment of the deltaic region, in Kolkata (*Kwan et al., 2013*) and the
479 Sundarban wetlands (*Binelli et al., 2007*). PCBs have been studied in sediment of ten cities and
480 towns located in the upper, middle and lower reaches of the main channel (Haridwar, Kanpur,
481 Allahabad, Buxar, Patna, Mokama, Sultanganj, Kahalgaon, Rajmahal, Farakka) (*Senthilkumar et*
482 *al., 1999*), as well as in Delhi, Sundarban and the lower Hugli (*Binelli et al., 2009; B. Kumar et al.,*
483 *2012a*). PCBs in river water have been investigated only along the Yamuna in NCT (*B. Kumar et*
484 *al., 2012b*), and in irrigation canals, lakes ponds and drains in the region surrounding Delhi (*S.*
485 *Kumar et al., 2012*). The distribution of OTCs has also been investigated in water and sediment of
486 the Kolkata harbour, the lower Hugli and Sundarban (*Antizar-Ladislao et al., 2011; Garg et al.,*
487 *2011*), and in water along the main channel and three minor tributaries (Loni, Pandu and Ganda
488 Nala), in the Kanpur-Unnao region (*Ansari et al., 1998*).

489

490 **6.2 Occurrence of ICs**

491

492 The concentrations of industrial chemicals both in Ganga water and sediment exhibit a wide range
493 of variability, from less than one to hundreds of ng/L and from less than one to hundreds of µg/kg,
494 respectively. PAHs concentrations in some instances reach up to thousands of ng/L and µg/kg
495 respectively with the highest concentrations of acenaphthylene in water (MRV: 65850 ng/L,
496 Lucknow (*Malik et al., 2004*)) and benzo[a]anthracene in sediment (MRV: 5950 µg/kg d.w., Dehli
497 (*Agarwal et al., 2006*)).

498 The highest concentration of PBDEs in sediment was recorded for PBDE-47 (maximum value:
499 8.832 µg/kg d.w., Sundarban (*Binelli et al., 2007*)), but no measurements were made in water .

500 For PCBs, compounds with the highest concentration were PCB-18 in water (MRV: 314 ng/L,
501 Delhi (*S. Kumar et al., 2012*)) and PCB-44 in sediment (MRV: 14.17 µg/kg d.w., Delhi (*S. Kumar*
502 *et al., 2012*)). The highest water concentration of OTCs in water was recorded for dibutyltin (DBT,
503 MRV: 104 ng Sn/L, Kolkata (*Garg et al., 2011*)), with the highest sediment concentration for TBT
504 (MRV: 442 ng Sn/g d.w., Kolkata (*Garg et al., 2011*)).

505

506 With regard to the spatial distribution of ICs, no clear trend could be detected along the main Ganga
507 channel, although *Binelli et al. (2009)* detected higher concentrations of PCBs in sediment within
508 the delta region (up to 26.84 µg/kg d.w.), compared to those found ten years earlier along the main
509 channel (ranging from 0.9 to 9.4 µg/kg d.w., (*Senthilkumar et al., 1999*)). This was attributed to
510 local point sources of pollution in the delta arising from urban sewage (*Binelli et al. 2009*)).

511 Sediment concentrations of OTCs detected by *Garg et al. (2011)* in Kolkata harbour were also one
512 order of magnitude higher than those detected in Sundarban and the lower Hugli reach (*Antizar-*
513 *Ladislao et al., 2011*), resulting from more limited water exchange in the harbour and direct sources

514 of antifouling paints. In surface water, concentrations appeared to be higher in Kolkata harbour
515 (*Garg et al., 2011*) than the Kanpur-Unnao region (*Ansari et al., 1998*) and shows that despite the
516 gradual decrease in organotin-based paints, contamination levels were high until recently.
517 Moreover, Kolkata is a contamination hotspot of PBDEs (*Kwan et al., 2013*).

518

519 For PAHs in the Hugli reach, concentrations reported by *Zuloaga et al. (2013)* were systematically
520 higher than those found by *Guzzella et al. (2005)* that were below detection limits at most sites.
521 This increasing trend is attributed to the presence of local point inputs from industrial sources and
522 other combustion processes (*Zuloaga et al., 2013*). All the studies on the Gomti, however, (*Malik et*
523 *al., 2011, 2004; Tripathi et al., 2009*) reported comparable although highly variable concentrations,
524 with sediment-associated total PAHs ranging from 50 µg/kg d.w. to more than 3000 µg/kg d.w.
525 Surprisingly high concentrations of total PAHs were detected by in the Himalayan region of
526 Nainital lakes, attributed to frequent forest fires and the use of coal and wood for heating and
527 cooking purposes (*Choudhary and Routh, 2010*). This would appear to be a recent phenomenon as
528 publications in the mid-1990s recorded very low PAH concentrations (*Ahmad et al., 1996*).

529

530 **7. Health and environmental risks of organic contaminants in the Ganga** 531 **compared with other river systems in India and worldwide**

532 **7.1 Emerging contaminants**

533

534 *Sharma et al. (2019)* reported maximum concentrations of many PPCPs (e.g. the NSAIDs
535 diclofenac, ibuprofen, naproxen) lower than those found in Kaveri, Vellar and Tamiraparani rivers
536 in peninsular India (*Shanmugam et al., 2014*), which are similar or even lower than those detected
537 by *Mutiyar et al. (2018)* in NCT and by *Williams et al. (2019)* in Udaipur (Ahar river).
538 Concentrations of ciprofloxacin in the Ganga were up to six orders of magnitude lower than those
539 found in the Isakavagu-Nakkavagu rivers in Hyderabad (*Fick et al., 2009*). Similarly, values of
540 triclosan reported from southern India, in the Tamiraparani, Kaveri and Vellar rivers (*Ramaswamy*
541 *et al., 2011*), were higher than those found in the Ganga, but comparable to those found by *Nag et*
542 *al. (2018)* in the Gomti. Reference values are reported in **table S1**.

543 At present it is not possible to determine the impact of these concentration levels on ecosystem
544 health because of the lack of official regulation or guidelines for ECs. Nevertheless, ecological risk
545 assessments performed by *Sharma et al. (2019)* and *Mutiyar et al. (2018)* demonstrated that the
546 detected values of PPCPs along the main Ganga channel and in Delhi posed no significant human
547 health concern, although there was a moderate risk for aquatic organisms (algae and *Daphnia*
548 *magna*) associated with some of the PPCPs. Neither the most detected PPCPs (i.e. acetaminophen,
549 carbamazepine, ibuprofen, ketoprofen, sulfamethoxazole and DEET) nor caffeine, whose
550 concentrations were the highest recorded among ECs, exceeded the predicted no-effect
551 concentrations (PNECs) for invertebrates and fish summarized by *Sharma et al. (2019)* and *Mutiyar*
552 *et al. (2018)*. However, caffeine, sulfamethoxazole and triclosan exceeded the PNECs calculated for
553 algae (15, 27 and 1.4 ng/L, respectively).

554 According to *Mutiyar et al. (2018)*, antibiotic residues detected in Delhi were concentrations shown
555 to cause acute toxicity; in particular, maximum concentrations of ciprofloxacin (1190 ng/L)

556 approached those causing growth inhibition to algae. It remains to be seen whether these
557 compounds exhibit synergic effects in case of exposure to multiple active substances. Besides acute
558 toxicity, antibiotics pose the risk of antimicrobial resistance, which has been extensively recorded in
559 bacterial isolates recovered from Indian surface waters, including the Gomti, the Yamuna and the
560 Kshipra rivers (*Diwan et al., 2018; Philip et al., 2018*). In the case of triclosan, all the
561 concentrations reported by *Nag et al. (2018)*, despite posing no risk for human health, exceeded the
562 Canadian Federal Environmental Quality Guideline (FEQG, 0.47 µg/L, **table S3**), a reference value
563 expressing the likelihood of direct adverse effects on aquatic life. For PFAS, according to *Yeung et al. (2009)*
564 river water concentrations from India appeared to be lower than those reported for other
565 Asian countries. In India, higher concentrations of PFOS and PFOA were found in the Cooum
566 River in Chennai and in Sri Lanka (**table S1**). Sediment concentrations of PFOS recorded in the
567 Hugli estuary were low, being below the detection limit in all the sampling stations. PFOA
568 concentrations are however comparable to those recorded in river estuaries of the Bohai Bay, China,
569 and one-two orders of magnitude higher than those recorded in Vietnam (*Lam et al., 2017*). PFOA
570 and PFOS were below the WHO drinking water guidelines (4 and 0.4 µg/L respectively, **table S3**),
571 and PFOS was below the Canadian FEQG (6.8 µg/L, **table S3**). With reference to other classes of
572 ECs, the sediment concentration of phthalates analysed in the Gomti were lower in comparison to
573 the values recorded in China and Taiwan (especially di-(2-ethylhexyl) phthalate (*Srivastava et al.,*
574 *2010*)), while water concentrations of benzotriazole and bisphenol A were comparable to those
575 found in other Asian countries (hundreds of ng/L (*Williams et al., 2019*)). Bisphenol A was below
576 the Canadian FEQG (3.5 µg/L, **table S3**).

577

578 7.2 Pesticides

579

580 Despite multiple restrictions of pesticide use (*Agarwal et al., 2015*) very high levels of pesticides
581 are still detected in the Ganga basin. *Mondal et al. (2018)* reported residues of α -, β - and δ -HCH
582 and p,p'-DDT exceeding the EC limit in drinking water (0.10 µg/L for single pesticide, **table S3**) in
583 56.2 and 100 per cent of samples collected in river water samples of the delta region, while only 6.2
584 and 12.5 per cent of samples were above the EC limit for the 16 detected pesticides (DDT and
585 metabolites, HCH isomers, endosulfan isomers, methylparathion, monocrotophos, phorate,
586 butachlor). Concentrations exceeding EC drinking water limits have also been reported along the
587 Ganga, in Allahabad (*Raghuvanshi et al., 2014*) and the Hugli (*Mondal et al., 2018*). In the case of
588 pond water collected in the delta region, the EC limit was exceeded in 25 per cent of methyl
589 parathion, 31.2 per cent of chlorpyrifos and 6.2 per cent of phorate and atrazine samples (*Mondal et al., 2018*).
590 Even higher water levels of α -HCH, α -endosulfan, dicofol, heptachlor, p,p'-DDE
591 (dichlorodiphenyldichloroethylene), alachlor and butachlor were found by (*Trivedi et al., 2016*) in
592 the Gomti river. Concentrations of α -HCH, aldrin and endosulfan found by (*Mutiyar and Mittal,*
593 *2013*) in Uttar Pradesh and Bihar exceeded the Indian drinking water quality standards (**table S3**).
594 Based on the assessment conducted by (*Mondal et al., 2018*), persistent OC pesticides such as HCH
595 isomers, DDT isomers and metabolites and endosulfan, still pose a potential risk to freshwater
596 aquatic animals and invertebrates.

597

598 7.3 Industrial compounds

599

600 *Malik et al. (2011)* found that total PAH concentrations in water and sediment of the Gomti are
601 higher than in other Asian rivers such as the Gao-ping in Taiwan, the Yellow River and the
602 Qiantang in China. However, the levels of PAHs in the Gomti appeared to be considerably lower
603 than that reported in the Jinsha river of China. *Choudhary and Routh (2010)*, who assessed the
604 impact of PAH pollution in sediment, found that fluorene, acenaphthylene and total PAH
605 concentrations exceeded the toxicological endpoints for aquatic fauna even in the Himalayan
606 districts of Nainital and Bhimtal. Similarly, *Zuloaga et al (2013)*, who analysed the distribution of
607 PAHs in the sediment of Sundarban wetlands, reported possible biological impact associated with
608 the recorded levels of pollutants. PBDEs sediment concentrations, especially those recorded in
609 Kolkata, are comparable to the PBDE levels found in other Asian areas (*Binelli et al., 2007*).

610 Maximum sediment concentrations of PCBs recorded in Sundarban and Delhi were higher than
611 those detected in the west coast of Sri Lanka (*Rajendran et al., 2005*) and in the southern part of the
612 Bay of Bengal (*Guruge and Tanabe, 2001*), whereas water concentrations were comparable to those
613 found in coastal waters of Daya Bay, China (*Zhou et al., 2001*). OTC concentrations reported in the
614 Ganga basin were generally lower than those of the coastal areas of Thailand and India (in
615 particular TBT) and in the sediment of the Zuari estuary, located on the west coast of India (*Garg
616 and Bhosle, 2005; Harino et al., 2006*). *Garg et al. (2011)*, however, observed that concentrations
617 of TBT compounds in water at all the sampling sites in Kolkata were higher than those known to
618 induce imposex in gastropods (< 10 ng/L). Reference values are reported in **table S2**.

619 Total PCB water values recorded in the Ganga basin were above both the Indian Drinking Water
620 Guidelines and the US EPA Water Quality Criteria (WQC, 0.5 and 0.14 $\mu\text{g/L}$ for acute and chronic
621 toxic effects on biota) (**table S3**). PCB levels in sediment were all below the Canadian Sediment
622 Quality Guideline (CSQG) for the protection of aquatic life (34.1 $\mu\text{g/kg}$ for total PCBs, **table S3**).

623 Concentrations of OTCs and TBT in water were all below the US WQC for both acute and chronic
624 toxic effects on biota (0.46 and 0.072 $\mu\text{g/L}$, respectively, **table S3**). But at many sites TBT
625 exceeded the European Environmental Quality Standards (EQS), both the Annual Average (AA,
626 0.0002 $\mu\text{g/L}$) and the Maximum Allowable Concentration (MAC, 0.0015 $\mu\text{g/L}$), representing
627 concentrations considered to protect the living environment against chronic and acute toxicity,
628 respectively (**table S3**). TBT concentration in sediment was higher than the upper guideline of the
629 Australian Sediment Quality Guideline Values for TBT (70 ng Sn/g, corresponding to around 29
630 $\mu\text{g/kg}$).

631

632 **Table 1.**

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634 **Table 2.**

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636 **Table 3.**

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638 **Table 4.**

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640 **Table 5.**

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642 **Table 6.**

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8. Research gaps and basin-scale implications

The review of 61 papers addressing surface water and sediment pollution revealed the presence of numerous knowledge gaps, the identification of which is essential for guiding future research campaigns and risk assessments.

One of the most critical research gaps is the lack of basin-scale assessments. No studies of this kind have been published either for ECs or ICs, whereas the first catchment review of pesticides was undertaken in 2013 (*Mutiyar and Mittal, 2013*). All earlier publications monitored either specific tributaries or the main channel, occasionally including canals or minor rivers. Given the size of the catchment and significant hydrologic, demographic and environmental variability in the region, only basin-scale studies will allow an understanding of the impact of tributaries on pollution patterns.

Further research is required also to understand the variability of concentrations according to the season (dry, winter and monsoon season) and the flooding regime. While only 13 papers (**tables S4-9**) provided an analysis of the seasonal variability of concentrations according to the river flow, studies of this kind would help the risk assessment by identifying the time frames in which higher concentrations are detected.

In the case of ECs, the gaps are exacerbated by the limited number of available studies, with only three publications focused on the cities along the Ganga main channel (*Sharma et al., 2019, 2016; Yeung et al., 2009*) and three on the NCT in the Yamuna sub-catchment (*Mutiyar et al., 2018; Mutiyar and Mittal, 2014a, 2012*), where the high population density is likely to constitute a direct source of PPCP contamination from domestic effluents. Although PPCPs and PFAS concentrations in the main channel were generally below 10 ng/L, and often close to detection limits, the widespread use of these compounds in densely populated areas, and their detection at higher concentrations in other regions of India (*Philip et al., 2018*), justify the necessity of further studies addressing the topic. Considering a resource-limited scenario, the most frequently detected compounds, such as acetaminophen, carbamazepine, ibuprofen, ketoprofen, sulfamethoxazole, DEET and caffeine, should be prioritized. Also antibiotics and antibiotic resistance, representing a major challenge for human health, should be further investigated.

Another knowledge gap is the lack of studies on pesticides other than OCPs and OPhs. While OCPs, for the most part, can be considered legacy compounds, no publications are available on carbamates and only one on pyrethroids, that include many active substances currently used in India (*Centre for Science and Environment, 2013*). Besides insecticides, also herbicides and fungicides have been poorly investigated.

9. Conclusions

This review demonstrates that data on organic contaminants in the Ganga basin is still fragmentary and mainly focused on the main channel, the Yamuna, the Gomti and the delta region.

685 The most studied organic contaminants were OCPs, followed by OPhs and PCBs. With reference to
686 ECs, the investigation of PPCPs has been particularly neglected in sediment, but widely
687 investigated in the case of ICs and, to a lesser extent, pesticides. Although pesticide concentrations
688 decreased between 1980 and 2019 as a result of restriction in their use, higher concentrations were
689 reported for PCBs and OTCs in the last decade. Recently hotspots of contamination have emerged
690 within and downstream of many of the large urban areas such as Delhi, Kanpur, Allahabad,
691 Varanasi, Patna, Kolkata, along the Gomti and in the Sundarban Wetlands. In these locations high
692 levels of all categories of pollutants have been reported with domestic and industrial effluents as
693 major contributors to pollution. Even pesticides, whose main source is agriculture, were often
694 reported in association with urban wastewater, since the two most studied insecticides, DDT and
695 HCH, have long been utilised for sanitation purposes in the region.

696
697 We recommend that future assessments should prioritize investigating ECs, especially PPCPs. For
698 pesticides, more focus is required for herbicides and carbamate insecticides that hitherto have not
699 been fully investigated. The seasonal variability of organic contaminants especially in relation to
700 flooding regime needs also to be studied.

701 The primary knowledge gap is a catchment-scale understanding of organic contaminant dispersal
702 and storage, including tributary contributions and downstream attenuation patterns in the main
703 Ganga channel. This is urgently needed for effective pollution control, watershed management and
704 the protection of human and ecosystem health.

705

706 **Acknowledgements**

707

708 This study was designed and financially supported by the Lincoln Centre for Water and Planetary
709 Health, University of Lincoln (UK). The work is also part of the activities planned within the
710 trilateral MoU agreement signed in 2017 by University of Padova, University of Lincoln and
711 Massey University.

712

713 **Author Contributions**

714

715 Conceptualization M.G.M.; Analysis and data interpretation A.G., M.K.R., A.M., M.G.M, R.M.,
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717 Editing A.G., A.M., M.G.M., P.T., R.M.; Funding Acquisition M.G.M.; Supervision P.T.

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719 **Declaration of Interests**

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721 Paolo Tarolli is a member of the iScience Editorial Board.

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723 **References**

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1019 **Figure 1. Study area and state boundaries within the Indian section of the Ganga basin.**

1020

1021 **Figure 2. Spatial distribution of the studied districts within the Ganga basin.**

1022 (A) Emerging contaminants in surface water and sediment. (B) Pesticides in surface water and sediment. (C) Industrial
1023 compounds in surface water and sediment.

1024

1025 **Figure 3. Maximum water concentrations of two selected ECs in the Ganga basin.**

1026 (A) Carbamazepine. (B) Sulfamethoxazole.

1027

1028 **Figure 4. Comparison between total DDT water concentrations in the 1990s (1990-1999) and the 2010s (2010-
1029 2019).**

1030 Total DDT stands for the sum of both o,o' -and p,p' - DDT, DDD and DDE.

1031

1032 **Figure 5. Comparison between total HCH water concentrations in the 1990s (1990-1999) and the 2010s (2010-
1033 2019).**

1034 Total HCH stands for the sum of α -, β -, γ - and δ -HCH.

1035

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1037 **Table S4. Main findings of papers addressing emerging contaminants in Ganga basin surface water. Related to
1038 table 1.**

1039 Abbreviations are listed in table S10.

1040

1041 **Table S5. Main findings of papers addressing emerging contaminants in Ganga basin river sediment. Related to
1042 table 2.**

1043 Abbreviations are listed in table S10.

1044

1045 **Table S6. Main findings of papers addressing pesticides in Ganga basin surface water. Related to table 3.**

1046 Abbreviations are listed in table S10.

1047

1048 **Table S7. Main findings of papers addressing pesticides in Ganga basin river sediment. Related to table 4.**

1049 Abbreviations are listed in table S10.

1050

1051 **Table S8. Main findings of papers addressing pesticides in Ganga basin surface water. Related to table 5.**

1052 Abbreviations are listed in table S10.

1053

1054 **Table S9. Main findings of papers addressing industrial compounds in Ganga basin river sediment. Related to
1055 table 6.**

1056 Abbreviations are listed in table S10.

1057

1058 **Table S10. List of acronyms and abbreviations reported in the tables. Related to tables 1-6 and tables S1-9.**

Tables

Table 1. Summary table of compounds, study areas and maximum concentrations of emerging contaminants in Ganga basin surface water.
Abbreviations are listed in **table S10**. See also **table S4**.

Compounds	Study area	Maximum concentration	ng/L	References
PFAS (20 compounds)	Ganga, Hugli	PFHxA	2.29	(Yeung et al., 2009)
Anionic surfactants	Hugli and small tributaries (Kolkata)	Total anionic surfactants	425,000	(Ghose et al., 2009)
NSAIDs, other pharmaceuticals	Yamuna (Delhi area)	-	-	(Mutiyaar and Mittal, 2012)
Other compounds (caffeine)	Yamuna (Delhi area)	Caffeine	808	Mutiyaar and Mittal, 2012)
Antibiotics	Yamuna (Delhi area)	Ampicillin	27,100	(Mutiyaar and Mittal, 2014a)
PFAS (21 compounds)	Bhagirathi, Alaknanda and Ganga	PFBS	10.2	(Sharma et al., 2016)
NSAIDs, other pharmaceuticals	Yamuna (Delhi area)	Ibuprofen	2302	(Mutiyaar et al., 2018)
Other compounds (caffeine)	Yamuna (Delhi area)	Caffeine	2640	(Mutiyaar et al., 2018)
Antibiotics	Kshipra (Ujjain)	Sulfamethoxazole	4660	(Diwan et al., 2018)
Biocides (triclosan)	Gomti	Triclosan	9650	(Nag et al., 2018)
Antibiotics, NSAIDs, other pharmaceuticals	Bhagirathi, Alaknanda and Ganga	Ketoprofen	107	(Sharma et al., 2019)
Insect repellent products, Biocides (DEET, triclocarban, triclosan)	Bhagirathi, Alaknanda and Ganga	DEET	22.3	(Sharma et al., 2019)
Artificial sweeteners	Bhagirathi, Alaknanda and Ganga	Saccharine	85.43	(Sharma et al., 2019)
Other compounds (caffeine)	Bhagirathi, Alaknanda and Ganga	Caffeine	743	(Sharma et al., 2019)
Antibiotics, NSAIDs, other pharmaceuticals	Ahar, Pichola Lake and Fateh Sagar Lake (Udaipur)	Caffeine	37,476	(Williams et al., 2019)
Hormones	Ahar, Pichola Lake and Fateh Sagar Lake (Udaipur)	Androsterone	1557	(Williams et al., 2019)
Insect repellent products, Biocides (DEET, triclocarban, triclosan)	Ahar, Pichola Lake and Fateh Sagar Lake (Udaipur)	DEET	388	(Williams et al., 2019)
Other compounds (bisphenol A, benzotriazole, methylbenzotriazole, caffeine)	Ahar, Pichola Lake and Fateh Sagar Lake (Udaipur)	Caffeine	37,476	(Williams et al., 2019)

Table 2. Summary table of compounds, study areas and maximum concentrations of emerging contaminants in Ganga basin river sediment.
Abbreviations are listed in **table S10**. See also **table S5**.

Compound	Study area	Maximum concentration	µg/kg d.w.	References
Phtalates	Gomti	DEHP	324.72	(Srivastava et al., 2010)
PFAS (PFOA, PFOS)	Hugli, Sundarban wetland	PFOA	14.09	(Corsolini et al., 2012)
Antibiotics	Kshipra (Ujjain)	Ofloxacin	9.74	(Diwan et al., 2018)
Biocides (triclosan)	Gomti	Triclosan	50.35	(Nag et al., 2018)
NSAIDs, other pharmaceuticals	Hugli	Carbamazepine	519	(Chakraborty et al., 2019)
Biocides (triclosan), Musk fragrances, Preservatives (parabens)	Hugli	Methyl paraben	423	(Chakraborty et al., 2019)
Other compounds (bisphenol A, phtalates, DEHA)	Hugli	DEHP	300	(Chakraborty et al., 2019)

Table 3. Summary table of compounds, study areas and maximum concentrations of pesticides in Ganga basin surface water.
Abbreviations are listed in **table S10**. See also **table S6**.

Compound classes	Study area	Maximum concentration	ng/L	References
OCPs	Yamuna (Delhi area)	p,p'-DDT	1610	(Agarwal et al., 1986)
OCPs	Mahala water reservoir (Jaipur)	γ -HCH	26,360	(Bakre et al., 1990)
OCPs	Yamuna (Delhi area)	Dieldrin	100,000	(Nair et al., 1991)
OCPs	Ganga (Varanasi)	p'-DDT	79,818	(Nayak et al., 1995)
OCPs; Herbicides; OPhs	Ganga (Kachla to Kannauj)	p,p'-DDT	5330	(Rehana et al., 1995)
OCPs; Herbicides; OPhs	Ganga (Narora)	α -HCH	1380	(Rehana et al., 1996)
OCPs	22 ponds (Shahjahanpur)	β -HCH	10,110	(Dua et al., 1996)
OCPs	7 Himalayan lakes (Nainital region)	p,p'-DDT	22,222	(Dua et al., 1998)
OCPs	Rivers and streams of the Kumaun Himalayan region	Total DDT	90 72	(Sarkar et al., 2003)
OCPs; OPhs	Ganga (Kanpur)	Malathion	2610	(Sankararamakrishnan et al., 2005)
OCPs; Herbicides; OPhs	Yamuna (Delhi area)	Total endosulfan	114	(Aleem and Malik, 2005)
OCPs	Bhagirathi, Alaknanda, Ganga and minor rivers of Uttarakhand	Total DDT	364.81	(Semwal and Akolkar, 2006)
OCPs	Streams, ponds and canals between Kanpur and Lucknow	β -HCH	1320	(Singh et al., 2007)
OCPs	Yamuna and canals (Delhi and Haryana)	p,p'-DDT	1423.44	(Kaushik et al., 2008)
OCPs	Gomti	β -HCH	301.44	(Malik et al., 2009)
OCPs	Hugli and small tributaries (Kolkata)	Other HCH isomers	7820	(Ghose et al., 2009)
OCPs; OPhs; Pyrethroids	flowing water bodies adjacent to the tea gardens of Dooars and Hill regions	Heptachlor	7.6	(Bishnu et al., 2009)
OCPs	Sharda river, Reetha river, drains surrounding lindane factory (Lucknow)	α -HCH	290,000	(Jit et al., 2011)
OCPs; OPhs	Ganga and Jamania river (Bhagalpur)	α -endosulfan	739	(Singh et al., 2012)
OCPs	Yamuna (Delhi area)	p,p'-DDT	239	(B. Kumar et al., 2012b)
OCPs	Ganga and tributaries in upper, middle and lower reach	Total endosulfan	17.9	(Mutiyaar and Mittal, 2013)
OCPs	Ganga and Yamuna (Allahabad)	γ -HCH	24,500	(Raghuvanshi et al., 2014)
OCPs; OPhs	Tighra reservoir (Gwalior)	Dichlorvos	22.3	(Rao and Wani, 2015)
OCPs; Herbicides	Gomti	Buthachlor	135,000	(Trivedi et al., 2016)
OCPs; OPhs; Herbicides; Fungicides	Hugli	δ -HCH	2940	(Mondal et al., 2018)

Table 4. Summary table of compounds, study areas and maximum concentrations of pesticides in Ganga basin river sediment.Abbreviations are listed in **table S10**. See also **table S7**.

Compound classes	Study area	Maximum concentration	µg/kg d.w.	References
OCPs	Yamuna (Delhi area)	p,p'-DDT	3060	(Agarwal et al., 1986)
OCPs	22 ponds (Shahjahanpur)	o,p'-DDT	908.25	(Dua et al., 1996)
OCPs	Ganga (Narora to Kannauj)	Heptachlor epoxide	18	(Ahmad et al., 1996)
OCPs	Ganga and tributaries in upper, middle and lower reach	Chlordane+metabolites	49	(Senthilkumar et al., 1999)
OCPs	Hugli	Endosulfan sulfate	400	(Bhattacharya et al., 2003)
OCPs	Hugli, Sundarban wetland	p,p'-DDT	1.29	(Guzzella et al., 2005)
OCPs	Bhagirathi, Alaknanda, Ganga and minor rivers of Uttarakhand	Not detected	-	(Semwal and Akolkar, 2006)
OCPs	Hugli, Sundarban wetland	p,p'-DDT	8.48	(Sarkar et al., 2008)
OCPs	Gomti	o,p'-DDT	345.66	(Malik et al., 2009)
OCPs	Yamuna (Delhi area)	Endrin aldehyde	90.87	(Pandey et al., 2011)
OCPs	Drains discharging into Yamuna (Delhi area)	Chlorpyrifos	286.56	(Kumar et al., 2011)
OCPs; OPhs	Ganga and Jamania River (Bhagalpur)	p,p'-DDT	3329.3	(Singh et al., 2012)
OCPs	Wetlands in Keoladeo National Park	γ-HCH	7540	(Singh Bhadouria et al., 2012)
OCPs	Ganga and Yamuna (Allahabad)	γ-HCH	19.8	(Raghuvanshi et al., 2014)
OCPs; OPhs; Herbicides; Fungicides	Hugli	δ-HCH	0.987	(Mondal et al., 2018)

Table 5. Summary table of compounds, study areas and maximum concentrations of industrial compounds in Ganga basin surface water.Abbreviations are listed in **table S10**. See also **table S8**.

Compound classes	Study area	Maximum concentration	ng/L	References
OTCs (dimethyltin, monobutyltin, dibutyltin, tributyltin)	Ganga, Pandu, Loni and Ganda Nala rivers (Kanpur-Unnao)	MBT	70.1 (ng Sn/L)	(Ansari et al., 1998)
PAHs (16 compounds)	Gomti	Acenaphthylene	65,850	(Malik et al., 2004)
PAHs (16 compounds)	Gomti	Acenaphthylene	82,670	(Malik et al., 2011)
OTCs (monobutyltin, dibutyltin, tributyltin)	Kolkata harbor	DBT	104 (ng Sn/L)	(Garg et al., 2011)
PCBs (28 congeners)	Yamuna and canals, lakes, ponds and drains (Delhi area)	PCB-44	594	(S. Kumar et al., 2012)
PCBs (27 congeners)	Yamuna (Delhi area)	PCB-18	280	(B. Kumar et al., 2012b)
PAHs (16 compounds)	Bhagirathi, Alaknanda and Ganga	Pyrene	21.21	(Sharma et al., 2018)

Table 6. Summary table of compounds, study areas and maximum concentrations of industrial compounds in Ganga basin river sediment.Abbreviations are listed in **table S10**. See also **table S9**.

Compound classes	Study area	Maximum concentration	µg/kg d.w.	References
PAHs (benzo[a]pyrene, phenantrene)	Ganga (Narora to Kannauj)	Phenantrene	18	(Ahmad et al., 1996)
Total PCBs	Ganga and tributaries in upper, middle and lower reach	Total PCBs	15	(Senthikumar et al., 1999)
PAHs (16 compounds)	Gomti	Benzo[a]anthracene+chrysene	1569.94	(Malik et al., 2004)
PAHs (19 compounds)	Hugli, Sundarban wetland	Fluoranthene	214	(Guzzella et al., 2005)
PCBs (13 congeners); PAHs (19 compounds)	Hugli, Sundarban wetland	PCB-153	0.54	(Guzzella et al., 2005)
PAHs (16 compounds)	Yamuna (Delhi area)	Naphtalene	4610	(Agarwal et al., 2006)
PBDEs (12 congeners)	Hugli, Sundarban wetland	PBDE-47	8.832	(Binelli et al., 2007)
Total PAHs (19 compounds)	Hugli, Sundarban wetland	Total PAHs	4249.71	(Binelli et al., 2008)
PCBs (23 congeners)	Hugli, Sundarban wetland	PCB-138	6.08	(Binelli et al., 2009)
PAHs (16 compounds)	Hugli, Sundarban wetland	Acenaphthylene	1521	(Tripathi et al., 2009)
Total PAHs (10 compounds)	Nainital and Bhimtal Lakes	Total PAHs	217,000	(Choudhary and Routh, 2010)
PAHs (16 compounds)	Gomti	Acenaphthylene	2726.4	(Malik et al., 2011)
OTCs	Hugli, Sundarban wetland	TBT	84.2	(Antizar-Ladislao et al., 2011)
OTCs	Kolkata harbour	TBT	442 (ng Sn/g)	(Garg et al., 2011)
PCBs (28 congeners)	Yamuna (Delhi area)	PCB-44	14.17	(B. Kumar et al., 2012b)
PBDEs (22 congeners)	Canals in Kolkata	PBDE-47	0.615	(Kwan et al., 2013)
PAHs (16 compounds)	Hugli, Sundarban wetland	Fluoranthene	1839.5	(Zuloaga et al., 2013)

 Ganga basin  India state boundaries



