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A review of the occurrence of pharmaceuticals and personal care products in Indian water bodies

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ABSTRACT

Little information exists on the occurrence and the ultimate fate of pharmaceuticals in the water bodies in India despite being one of the world leaders in pharmaceutical production and consumption. This paper has reviewed 19 published reports of pharmaceutical occurrence in the aquatic environment in India [conventional activated sludge wastewater treatment plants (WTPs), hospital WTPs, rivers, and groundwater]. Carbamazepine (antipsychoactive), atenolol (antihypertensive), triclocarban and triclosan (antimicrobials), trimethoprim and sulfamethoxazole (antibacterials), ibuprofen and acetaminophen (analgesics), and caffeine (stimulant) are the most commonly detected at higher concentrations in Indian WTPs that treat predominantly the domestic sewage. The concentration of ciprofloxacin, sulfamethoxazole, amoxicillin, norfloxacin, and ofloxacin in Indian WTPs were up to 40 times higher than that in other countries in Europe, Australia, Asia, and North America. A very few studies in Indian rivers reported the presence of ciprofloxacin, enoxacin, ketoprofen, erythromycin, naproxen, ibuprofen, diclofenac and enrofloxacin. Similar compounds were reported in rivers in China, indicating a similar usage pattern in both of these developing countries. In a study reported from an open well in southern India, the groundwater showed the presence of cetirizine, ciprofloxacin, enoxacin, citalopram and terbinafine, which was close to a WTP receiving effluents from pharmaceutical production.

1. Introduction

Pharmaceuticals and personal care products (PPCPs) include active ingredients of prescription and non-prescription drugs for human and veterinary use, disinfectants, illicit drugs, body lotions, etc. (Kaplan, 2013; Bu et al., 2013). The PPCPs thus consumed evoke a specific biological response from the host, after which are ultimately discharged into the environment. Hirsch et al. (1999) and Kummerer (2009) have reported that ~10–90% of the administered dose of PPCPs are excreted from the human body in their parent form, while the rest are excreted as metabolites and/or conjugated forms. The excreted PPCPs reach the wastewater treatment plants (WTPs) and finally discharge raw or treated effluent into the groundwater, rivers, lakes, oceans, and soil (Fig. 1). They have been detected in the aquatic environment since the 1970s (Veach and Bernot. 2011 and references therein), and in the last twenty years, in all types of surface water, groundwater and the oceanic environment (WHO, 2011; Klosterhaus et al., 2013; Luo et al., 2014). In the aquatic environment, PPCPs can be toxic to certain aquatic organisms and trigger antibiotic resistance amongst pathogens (Behera et al., 2011; Kidd et al., 2007; Xiao et al., 2001; Kolpin et al., 2002; Kristiansson et al., 2011). Nevertheless, limited literature exists for establishing the effects of a cocktail of PPCP mixture in the environment, on the aquatic biota and the humans (Tixier et al., 2003; Daughton and Ternes, 1999).

India is among the top five producers of pharmaceutical chemicals, with an expected turnover of USD 45 billion per year by 2020 (KPMG International, 2006). The organized sector of Indian pharmaceuticals consists of around 250–300 companies, with its drug exports growing 30% annually (KPMG International, 2006). In other words, every third pill taken in the world is manufactured in India. Among the bulk formulations, around 80% have been reported to be consumed indigenously (Kallummal and Bugalya, 2012). On the other hand, treatment capacity of domestic sewage in India is far below the quantity of sewage generated from 1.3 billion people; only 31% of the total sewage

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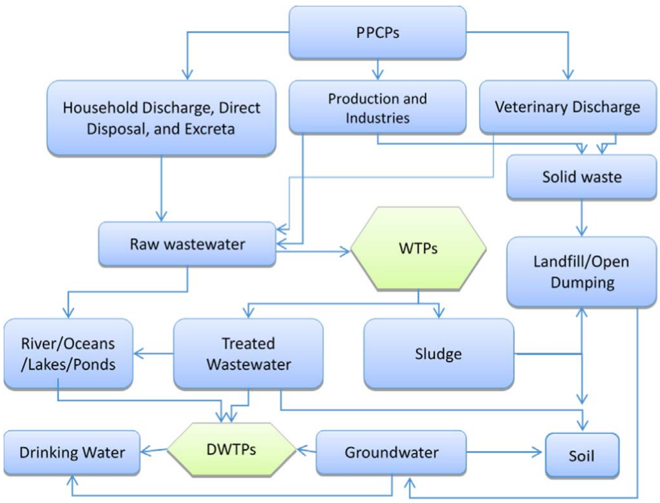

Fig. 1. Flow diagram of the PPCP pathways in the environment. WTPs: Wastewater treatment plants, DWTPs: Drinking water treatment plants.

produced (~38,254 million liters per day) in 908 cities were treated in 2008 (Subedi et al., 2015a).

Despite high rates of production and consumption of PPCPs across the country and shortage in demand and supply for the sewage treatment, limited literature is available to account for their occurrence, transport, and fate in the aquatic environment (Subedi et al., 2015a; Subedi et al., 2015b; Rehman et al., 2013; Mutiyar and Mittal, 2014). This review provides an overview of levels of PPCP contamination in Indian water bodies, which can potentially trigger more large-scale nationwide studies on the occurrence of PPCPs and their ecological impacts. The pharmaceutical residue levels in domestic wastewater, hospital effluent, river water, and groundwater in India are compared with that reported elsewhere. Finally, recommendations for an efficient management of PPCP contamination in the aquatic environment, that are important for the sustainable solution, are provided.

2. Pharmaceutical contaminants in India

2.1. Wastewater treatment plants

Twelve studies have reported the pharmaceuticals in wastewater from conventional activated sludge treatment based WTPs in India (Larsson et al., 2007; Fick et al., 2009; Mutiyar and Mittal, 2013a, 2014; Singh et al., 2014; Akiba et al., 2015; Subedi et al., 2015a; Prabhasankar et al., 2016; Archana et al., 2016; Mohapatra et al., 2016; Anumol et al., 2016; Subedi et al., 2017) (Table 1).

WTP outlets are the primary point sources of pharmaceutical contamination in the rivers and oceans (Daughton and Ternes, 1999). The existing wastewater treatment processes are incapable of removing most of the pharmaceutical contaminants; removal efficiencies typically ranged from 12.5% to 100% (Santos et al., 2007; Luo et al., 2014). The microbial transformation and/or deconjugation of glucuronides of the select pharmaceuticals and their active metabolites can have negative removal efficiency (Subedi et al., 2015a). Removal efficiency depends on the treatment process, sludge age, the geography of the area, and the rainfall rate (Chen et al., 2012). The overall pharmaceutical contamination profile is also dependent on the pharmaceutical production and usage pattern (Behera et al., 2011).

2.1.1. WTP receiving effluents from pharmaceutical industries

Despite of relatively lower levels (ng/L to µg/L) of pharmaceuticals in wastewater from WTPs that process predominantly domestic sewage, much higher concentrations (mg/L) of pharmaceutical contaminants were reported from the WTPs that process wastewater from the

Table 1
Mean reported concentrations of pharmaceuticals and their metabolites in wastewater (ng/L) from wastewater treatment plants (WTPs) in India.

Contaminants	Influent	Effluent
Antischizophrenics		
Quetiapine	38[a], 15[b], 36.8[k], 20.8[l], 24.8[m], 13.8[n], 71.2[o]	20[a], 5.2[b], 6.32[l], 16.6[m], 22.4[o]
Noquetiapine	1.87[k], 6.78[l], 4.70[m], 10.7[n], 16.4[o]	4.04[k], 10.1[m], 1.92[n], 6.50[o]
Aripiprazole	44[a], 29[b], 4.20[l], 14[m],	71[a], 0.4[b]
Dehydroaripiprazole	3.80[k], 0.90[l]	2.20[k]
Sedatives-hypnotics-anxiolytics		
Lorazepam	46[a],26[b], 23.6[n], 19.8[o]	23[a],12[b], 19.1[k], 27.4[l], 24.4[m], 8.26[n], 41.8[o]
Alprazolam	41[a], 10.1[k], 4.20[l], 6.98[o]	33[a], 25[b], 6.94[k], 5.72[l], 2.52[o]
α-hydroxyalprazolam		8.48[k]
Diazepam	23[a], 25[b], 6.80[k], 4.46[l], 6.66[n], 196[o]	36[a], 9.5[b], 8.20[k], 47.0[l], 24.6[n], 238[o]
Oxazepam	140[a], 50[b], 25.0[m], 13.7[o]	85[a], 50[b], 38.2[m], 17.0[n], 17.0[o]
Nordiazepam	12[a], 5.9[b], 11.4[k], 5.40[l], 14.5[m], 3.26[n], 12.4[o]	85[a], 50[b], 10.5[k], 6.70[l], 8.56[m], 3.08[n], 5.96[o]
Carbamazepine	450[a], 550[b], 470[d], 650[e], 5800[i], 8200[j], 82.2[k], 270[l], 840[m], 22.0[n], 726[o]	580[a], 480[b], 88[k], 236[l], 900[m], 147[n], 318[o]
Antidepressants		
Venlafaxine	38[a], 5[b], 30.6[k], 10.3[l], 138[m], 9.30[n], 46.2[o]	15[a], 5[b], 6.70[k], 7.96[l], 105[m], 7.26[n], 29.4[o]
Bupropion	19[a], 23[b]	14[a], 5[b], 3.80[k], 3.42[o]
Sertraline	23[a], 40[b], 5.33[k], 2.53[l], 87.0[m], 10.6[n], 21.8[o]	18[a], 1.7[b], 59.8[m], 10.8[o]
Nosertraline	116[k], 144[l], 386[m]	55.6[k], 57.6[l], 50.0[m]
Citalopram	7.16[l], 16.4[n], 31.8[o]	9.46[m], 14.7[n], 29.8[o]
Antihypertensives		
Propranolol	51[a], 43[b], 17.0[k], 18.5[l], 34.2[m], 14.5[n], 123[o]	43[a], 28[b], 7.98[k], 11.8[l], 37.6[m], 11.4[n], 12.3[o]
Atenolol	2900[a], 1400[b], 41400[i], 13800[j], 1010[k], 374[l], 2440[m], 192[n], 1910[o]	1500[a], 590[b],197[k], 244[l], 2500[m], 16.3[n], 772[o]
Metoprolol	35500[i], 11800[j]	
Diltiazem	55[a], 16[b], 5.64[n], 1.39[o]	5[a], 1.8[b], 1.52[m], 1.53[o]
Desacetyl diltiazem	32[a], 6.40[k], 1.04[l], 7.62[m], 1.55[n], 44.4[o]	44[a], 10[b], 3.02[k], 1.82[l], 8.96[m], 1.51[n], 20.0[o]
Verapamil	36[a], 25[b], 1.74[k], 0.74[l], 0.61[o]	2[a], 0.88[l], 1.08[m], 2.64[o]
Norverapamil	260[a], 47[b], 0.88[k], 4.04[m],	4[a], 1.46[m],
Antimicrobial		
Triclocarban	2400[a], 4000[b], 515[k], 933[l], 8880[m], 1150[n], 2100[o]	540[a], 260[b], 22.4[k], 457[l], 5860[m], 48.4[n], 375[o]
Triclosan	4890[f], 450[k], 145[l], 2500[m], 892[n], 2440[o]	3500[f], 2500[m], 202[n]
Antibiotics/fungicides		
Trimethoprim	180[a], 29[b], 4010[d], 210[e], 3[h], 4[h], 23[h], 33.0[k], 90.8[l], 156[m], 160[n], 35.6[o]	25[b], 8[h], 1[h], 3[h], 34.8[k], 38.0[l], 103[m], 2080[o]
Sulfamethoxazole	220[a], 100[b], 3[h], 66[h], 195[k], 288[l], 552[m], 414[n], 2260[o]	260[a], 25[b], 13[h], 27[h], 9[h], 70.2[l], 318[m], 228[n], 296[o]
Ampicilin	104.2[c]	12.68[c]
Ciprofloxacin	20.06[c], 12900[f]	8[c], 11670[f]
Erythromycin	12[h]	2[h], 1[h], 9[h]
Gatifloxacin	2.74[c]	1.22[c]
Levofloxacin	86700[i], 107900[j]	
Nofluoxacin	18200[i]	
Azithromycin	176900[i], 29300[j]	
Sparfloxacin	22.49[c]	0.14[c]
Cefuroxime	3.42[c]	0.22[c]
Ofloxacin		0–212[g]
Clindamycin	210[a], 31[b], 5.16[k], 18.3[l], 27.2[m], 49.6[n], 1870[o]	25[b], 48.0[k], 6.96[l], 17.5[m], 63.8[n], 952[o]

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