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EDITORIAL

Occurrence and Spread of Emerging Organic Pollutants and Antibiotic Resistance in Urban Waters of Sri Lanka

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Abstract: Emerging pollutants such as Pharmaceuticals and Personal Care Products (PPCPs) in water resources are now considered a global water quality issue. Concerns associated with the presence of PPCPs in the environment include abnormal physiological processes and reproductive impairment, increased cancer incidence, the development of antibiotic-resistant bacteria, and the potential for increased toxicity of chemical mixtures. However, the number of studies on PPCPs in the aquatic environment and their use in wastewater tracking in developing countries, such as Sri Lanka, remains limited. Consequently, during the last five years, we have conducted various research to establish the comparative occurrence of PPCPs and antibiotics in urban waters in Sri Lanka. Our studies detected around 20 PPCPs in urban waters in Sri Lanka. Acetaminophen was dominant in hospital discharge (up to 124 µg/L), while caffeine was the most significant contributor to municipal wastewater (up to 69 µg/L). We suggest using Caffeine, Carbamazepine, and Acetaminophen for detecting domestic pollution in groundwater and surface water. We noticed that 50 to 100 % of antibiotic resistance in urban waters of Sri Lanka is comparatively higher than that of other countries, alarming that multidrug-resistant has become a significant concern for community health.

Keywords*: Emerging pollutants, PPCPs, Antibiotic Resistance, Urban waters, Sri Lanka*

Introduction

According to the estimates and projections of the world urban and rural populations published by the United Nations, over half of the global population lives in urban areas. It is estimated that by 2050, the urban population will be 6.68 billion, compared to 3.09 billion in rural areas. The urbanization rate is two- to threefold more rapid in less developed regions than in developed areas (United Nations, 2019). Rapid urbanization often exceeds urban infrastructure development in developing countries, especially wastewater management (WHO, 2017). Poor wastewater management and outdated sanitation facilities continuously pose potential health issues and pollution problems for the aquatic environment in the developing context.

In recent decades, studies on wastewater characteristics have drawn attention towards the environmental occurrence of various newly identified compounds of anthropogenic origin. The occurrence of such trace compounds (primarily organic), known as the "emerging pollutants," and their harmful impact on both aquatic and terrestrial life forms as well as on human health is now an issue of concern among the scientists, engineers, and the general public as well (Gogoi, 2018). Those emerging contaminants in water resources are now considered a global water quality challenge. Due to a lack of legislation and understanding, these pollutants are not regulated or monitored. On the other hand, conventional wastewater treatment plants do not entirely remove them. Many of the United Nations' Sustainable Development Goals (SDGs) include reducing emerging contaminants and their discharge into water resources. So far, the emerging problems of those new contaminants have not been sufficiently addressed. Some scientists argue that these emerging pollutants may have a negative impact on the SDGs and even threaten their achievement (Domingo-Echaburu et al., 2021).

In large part, contaminants whose new origin and in question are derived from two broad categories; Pharmaceuticals and Personal Care Products (PPCPs)

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and Endocrine Disrupting Compounds (EDCs). Pharmaceuticals are chemicals formulated into medicines to treat illness or slow their onset, such as antibiotics, painkillers, beta-blockers, lipid regulators, etc. Personal care products include cosmetics, fragrances, detergents, insect repellants, sun-screen agents, skin anti-aging preparations, and disinfectants (Chaminda et al., 2013a). Once consumed, some pharmaceuticals are transformed during metabolism while others remain unchanged before all are excreted and urine and feces. Potential concerns from the environmental presence of these emerging contaminants such as PPCPs include abnormal physiological processes and reproductive impairment, increased incidences of cancer, the development of antibiotic-resistant bacteria, and the potential increased toxicity of chemical mixtures (Gogoi et al., 2018). It must be recognized that even though individual concentrations of any PPCP might be below, the combined concentrations from PPCP sharing a common mechanism of action could be significant (Daughton & Ternes, 1999). Upon discharge of treated sewage into receiving water bodies, residual PPCPs may be diluted or blended with contaminants from other discharge points, as well as runoff and seepage (Chaminda et al., 2013a).

Because there is a lack of relevant data on the effects, fate, and concentration levels of emerging pollutants such as PPCPs, it is difficult for governments or policymakers to regulate their use and manage the present concentration in the environment. A series of research on pharmaceuticals in groundwater, surface water, wastewater, and drinking water have been well reported in many developed countries (Tran et al. 2018). However, the number of studies on pharmaceuticals in the water environment and their application in wastewater tracking in developing countries, including Sri Lanka, is still limited (Honda et al., 2011, Chaminda et al., 2013b, Kumar et al. 2019, Do et al., 2021, Kumar et al. 2021). Since the quantity, type, and patterns of PPCPs usage are geographically varied, the level of PPCP occurrence in the aquatic environment may differ in Sri Lanka compared to other countries. Urban waters are becoming significant PPCP sources; however, their vulnerability to such emerging pollutants in urban waters of Sri Lanka is a question. Therefore, over the past five years, we have undertaken several research studies to,

1. determine the comparative prevalence of PPCPs and Antibiotics in Urban waters in Sri Lanka
2. use of PPCPs as markers to detect fecal pollution in groundwater and surface water
3. determine the prevalence of antibiotic-resistant bacteria (ARB), antibiotic resistance genes (ARG), and their seasonal variation

Some of our research outputs from those studies can be considered the first-ever research publications in this field regarding the Sri Lankan context. We collected water samples at the influents and effluents from sewage treatment plants (STPs), hospital discharge, river waters (Kelani and Gin), and urban canals in Galle and Colombo. Samples were analyzed for PPCP parameters at the laboratories in Tokyo, Japan, using LC-MS/MS coupled to a triple quadrupole mass spectrometer. Water samples were pretreated at the University of Ruhuna, preserved, and kept frozen before transportation to Tokyo, Japan. The method quantitation limit for all analytes ranged from 0.005 to 0.15 ng/L (Do et al., 2021). We also tested for antibiotic resistance of *E. coli* bacteria in Kelani and Gin rivers, urban canals, and STPs in Galle and Colombo. The isolated *E. coli* colonies were evaluated for resistance to six antibiotics such as levofloxacin (LVX), ciprofloxacin (CIP), norfloxacin (NFX), kanamycin monosulphate (KM), tetracycline (TC), and sulfamethoxazole (ST). We used the Kirby-Bauer (KB) disc diffusion method (Honda et al., 2016) at the Environmental Engineering Laboratory, University of Ruhuna, Galle. Real-time quantitative reverse transcription PCR (qRT-PCR) was used to study the antibiotic resistance genes (ARG) and done at Kanazawa, Japan. Principal component analysis was performed to characterize the resistance patterns of the samples.

Prevalence of PPCPs in Urban Waters in Sri Lanka

We screened 82 PPCPs from sewerage treatment plants (STPs) and detected 20 pharmaceuticals. The concentrations of detected PPCP compounds in raw wastewater from the Hikkaduwa STP and hospital effluent (Karapitiya) were comparable with those frequently detected in previous studies in developed countries (Quyen et al., 2021). Acetaminophen (antipyretic) and Caffeine (stimulant) accounted for the highest total concentrations in both STP influent and hospital discharge. Carbamazepine (Neuroactive drugs), Cotinine (Tobacco metabolite), Sulfamethoxazole (Antibiotic), Sulfapyridine (Antibiotic), Atenolol (Beta-blocker), and Acesulfame (Artificial sweetener) were other detected PPCPs in raw sewerage and treated effluents of municipal STP and hospital effluent as well. Acetaminophen is a widely used over-the-counter

medicine and is often detected at high concentrations in domestic wastewater (Chaminda et al., 2013 a,b). The Acetaminophen concentration of raw sewage of Hikkaduwa STP was detected up to 43 µg/L, while Caffeine was found up to 69 µg/L. Despite the high concentrations, we found that Acetaminophen and Caffeine were generally removed up to 99% in the Hikkaduwa STP. Carbamazepine, Atenolol, Sulfapyridine, and Sulfamethoxazole were also detected up to 0.24, 0.48, 0.47 0.20 µg/L in municipal STP in Hikkaduwa. Unlikely Acetaminophen, it was observed that Carbamazepine and other detected PPCPs were not adequately removed during the biological treatment process in Hikkaduwa STP.

Acetaminophen is the highest concentrated PPCP found in hospital effluent and was detected up to 124 µg/L while two antibiotics (Sulfamethoxazole and Sulfapyridine) were detected up to 2.6 µg/L indicating the current treatment process used by the Karapitiya STP was not sufficient to remove pharmaceutical compounds. The complicated composition of hospital discharge suggests the need for a longer-term monitoring study. We also collected surface water samples from Galle municipal canals consisting of the Moragoda, Moda, and Kepu Ela, which also receive wastewater from residential areas. Our households survey found that two-thirds of the urban greywater is often discharged into above municipal canals in Galle.
We detected Acetaminophen and Caffeine up to 4.7 µg/L and 21.7 µg/L in urban canals within Galle city. Carbamazepine was also detected at low concentrations in those urban canals.

Further, we collected 32 water samples (31 private wells and one public well) during the 2019 monsoon season from groundwater wells in the Galle area. Four PPCPs at trace levels (ng/L) were detected in groundwater, suggesting wastewater impacts from the septic tanks. Caffeine was detected in 89% of the sample at a maximum concentration of 7.9 ng/L, indicating fecal pollution. Carbamazepine was detected in 42% of wells with a concentration of up to 6.9 ng/L, located close to and downstream of the septic tanks. We detected an antibiotic (Sulfamethoxazole) with concentrations as low as 1.1 ng/Lin groundwaters in the Galle area. We also found that Acetaminophen in groundwater was less than detectable, suggesting that physical and biological processes within the septic tanks and the saturated zone limit their transfer to the shallow groundwater (Do et al., 2021).

The detection of Caffeine indicated continuous wastewater discharge from poor sanitation systems. In contrast, the occurrence of persistent pollutants, such as Carbamazepine and Sulfamethoxazole, indicated the extent of pollutant transport from septic tanks. Therefore, we suggested that a setback distance >18m is unlikely to protect groundwater from septic tanks contamination. Thus, we recommend a more significant vertical separation from septic tanks to groundwater or the installation of watertight septic tanks where possible.

PPCPs as markers to detect domestic pollution in groundwater and surface water

Our studies suggest that Acetaminophen (antipyretic), Caffeine (stimulant), Carbamazepine (Neuroactive drugs), and Acesulfame (Artificial sweetener) can be used for identifying pollutant sources in Galle City. The stimulant, Caffeine, and the artificial sweetener, Acesulfame, are included in everyday beverages, such as tea, coffee, and soft drinks, so both black water and gray water may contain these compounds. However, the fact that Caffeine is readily degradable and entirely metabolized in the human body suggests that black water is not a significant source and is more likely to be a gray water indicator. However, due to the persistence of Acesulfame during wastewater treatment, it can be used to generalize the impacts of domestic wastewater on surface water. Acetaminophen (antipyretic) and Carbamazepine (Neuroactive drugs) are active pharmaceutical ingredients. As expected, the Carbamazepine level was significantly lower than that of Acetaminophen in urban wastewater owing to its limited use. Therefore, we suggested using Acetaminophen as an indicator for identifying the impact of untreated human excretion over short distances. Further, Carbamazepine could be a good indicator in tracking hospital residues over long distances due to its environmental persistence (Quyen et al., 2021). Our finding of the presence of PPCPs in well waters indicated the vulnerability of the drinking groundwater source to fecal contaminants from the outdated sanitation system. We observed that E.Coli and total coliform had a significant positive correlation with Caffeine concentration in groundwater. Moreover, a significant correlation was also observed between nitrate and Carbamazepine in groundwater (Do et al., 2021).

Resistance towards antibiotics

Our studies between October 2017 (wet season) and in March 2018 (dry season) evaluated the occurrence

of antibiotic-resistant bacteria (ARB) and antibiotic-resistant gene (ARG) for six antibiotics: Norfloxacin (NFX), Ciprofloxacin (CIP), Levofloxacin (LVX), Kanamycin monosulfate (KM), Tetracycline (TC), and Sulfamethoxazole (ST) in influents and effluent of sewerage treatment plant (STP) and surface waters in Colombo and Galle using KB disk diffusion method. Samples were collected from two municipal STPs from Colombo and hospital effluent from Galle. On the other hand, we collected water samples at different locations of Kelani River, Gin River, and some urban canals (Wellawatta Canals in Colombo, Moda Ela, and Moragoda Ela in Galle). For all STP samples, at least 50% of colonies showed resistance to all six antibiotics. Our antibiotic-resistant gene (ARG) screening test results were in agreement with the antibiotic resistance bacteria (ARB) test (Sovannlaksmy et al., 2020). Compared to municipal STP, the hospital effluent had a higher proportion of multidrug resistance due to the higher concentration of pharmaceutical products in the wastewater (Kumar et al., 2020 c). We noticed that no significant contribution of fecal contamination was found to increase the antibiotic resistance ratio, like previously reported studies. (Honda et al., 2015). *E. coli* prevalence was reduced during the treatment process, but the remaining bacteria could adapt in the presence of antibiotics, leading to a further increase in antibiotic resistance. Regarding some countries (India, Thailand, Poland, Ireland, Portugal, Austria, etc.), the STPs in Sri Lanka showed more antibiotic resistance and a consistent increase in antibiotic resistance after the treatment Kumar et al., 2020 c). *E. coli* bacteria isolated from the effluent of Jaela STP and hospital effluents from Galle (20 m and 50 m downstream) exhibited 100% of antibiotic resistance.

We further observed that almost all sampling points in Kelani and Gin Rivers and canals contained *E. coli* strains that showed resistance to more than four antibiotic multidrug resistance, alarming health concerns in the future. Our studies suggested high antibiotic resistance in the Kelani River (up to 90%) compared to the Gin River (up to 40%). Wellawatta Canals, Moda Ela, and Moragoda Ela showed more antibiotic-resistant *E. coli* than Gin and Kelani rivers owing to more pollution and lower flow rates in canals (Kumar et al., 2020 a). We found that the resistance percentage for older antibiotics like Sulfamethoxazole (ST) > Tetracycline (TC) was higher than the newer antibiotics in the Kelani River. Further, for non-fluoroquinolone (KM, ST, and TC), higher resistance was observed at downstream locations in Kelani River compared to upstream locations, which probably indicates the antibiotic use pattern affecting the resistance. On the other hand, the resistance percentage of the fluoroquinolone (LVX, CIP, and NFX) decreased from upstream to downstream in the Kelani River (Kumar et al., 2020 b).

Conclusion

Our studies detected around 20 PPCPs in urban waters in Sri Lanka. Acetaminophen was dominant in hospital discharge (up to 124 µg/L), while caffeine was the most significant contributor to STP influent (up to 69 µg/L). We suggest using Caffeine, Carbamazepine, and Acetaminophen for detecting domestic pollution in groundwater and surface water. We noticed that 50 to 100 % of antibiotic resistance in urban waters of Sri Lanka is comparatively higher than that of other countries alarming that multidrug-resistant has become a significant concern for community health. The study implies that urbanization, land use, and lifestyle are the three most critical factors governing multidrug resistance.

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