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Contaminants in Drinking and Wastewater Sources

Challenges and Reigning Technologies

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Dedicated
to
Research Life and Ethics
of
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Foreword

I am glad to announce that a book project entitled *Contaminants in Drinking and Wastewater Sources: Challenges and Reigning Technologies* is going to be published by Springer Nature Singapore Pvt. Ltd. under the dynamic lead editorship of Prof. Manish Kumar. The focal theme of this book is aptly chosen, as the widespread problems of water and environment pollution are jeopardizing our health. In this context, this multidisciplinary book focusing on actual challenges posed by contaminants of emerging concerns (CECs) on various compartments of the water cycle impacted by wastewater discharge and toxic leachates from waste disposal sites is the need of the hour. The main contribution of the present book is to aptly highlight a paradigm shift from conventional "priority" pollutants research to "emerging" or "new generation" unregulated contaminants study of the last 25 years duration. I am fully assured that the reader will get impressive and comprehensive information on structurally diverse groups of chemical compounds that have adverse effects on the aquatic environment, from this excellent meticulous compilation.

Another unique selling point of this book is that it not only defines the impacts of the environmental exposure of trace concentrations of CECs like pharmaceuticals, perchlorate, antibiotics, chlorophenol, microplastics, perfluorooctane-sulphonate and/or their metabolites, but also explores about the technological advancements. The research findings that are included in this book aptly explore the global strength, environmental status, chemical risk assessment and management strategies of CECs with relevant modern techniques, with the principle focus on concurrent "Emerging Water Quality Issues". My personal the most favourite component of this book is the discussion on the global human vulnerability due to the exposure of CECs in the natural environment that provides a unique platform for scientists from branches of science and technology including engineering sciences, agricultural sciences, biogeochemical sciences, hydrogeochemistry, toxicological sciences, social sciences, environmental policy and governance.

I also liked the chronology of the chapters which nicely flows through depicting monitoring and occurrence of CECs (distribution, source and fate in the aquatic environment) in its first part, and moves to advanced removal techniques of CECs (biotic, abiotic, photocatalytic degradation) in the second part for early prediction of risk and management purposes. Simultaneously, the present book also explores the human health perspectives of CECs, health risk assessment study along with case studies on sociological approaches. I hope this book will establish a road map for explaining in details the multifaceted application of biotic (bioreactor, anammox, etc.), abiotic (photocatalysis, ozonization, etc.) and natural removal system (constructed wetland) to be considered as effective treatment techniques for the emerging contaminants in the environment. I congratulate all the editors of the chapters for putting up a great work together.

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Preface

The widespread occurrence of water and environmental pollution is jeopardizing the health and sustainability of the ecosystems upon which all depend. Concerns about pollution have led people around the globe to focus their research on difference forms of contaminants. The changing research focus has resulted in paradigm shift from conventional “priority” pollutants to “emerging” or “new generation” mainly unregulated contaminants research over the last 25 years. Research findings presented in this book explore the global extent, environmental status, chemical risk assessment and management strategies for contaminants of emerging concern (CECs) with relevant modern techniques, with the principle theme of “Emerging Water Quality Issues.” This book is a meaningful integration of two major parts covering the subjects related to occurrence, fate and transformation products of emerging pollutants in aquatic environment, overview of water quality and risk study from aquatic toxicity, removal processes and wastewater treatment strategies social-ecological risk perspectives, challenges for regulation and management etc. Each part is comprised of at least seven chapters dealing with diverse topics, such as the life-cycle of emerging contaminants, human and eco toxicological risks posed by CECs, potential recommendations to diminish the effects of emerging contaminants, an update on biological treatment techniques, and sociological aspects. Various research and review papers along with case studies have been included in the present chapters to broaden the critical concept of researchers and engineers about the evaluation of risk assessment methodologies which in turn establish the proper functioning of wastewater treatment methodologies for the effective removal of CECs from various types of wastewater.

Part I of this book consists of seven articles dealing with the occurrence, persistence, metabolic transformations, and adverse human health effects of emerging contaminants in the environment and illustrates the state-of-the-art measurement (affinity-based methods) of detecting contaminants in the aqueous environment through advanced analytical techniques from ppm to ppb level. This part has also highlighted about the CECs of global/national importance such as nanomaterials (carbon nanotubes, graphene based, zeolites, nano composites) pharmaceuticals and personal care products (PPCPs) and artificial sweeteners (ASs), microplastics

(MPs), perchlorate (ClO4-), which are the representatives of different diverse regions across the globe starting from Southeast Asia, East Asia to South Asia. In the first chapter, the authors have critically evaluated and analyzed the anthropogenic biomarkers in Asian groundwater based on 23 studies conducted in six countries (China, India, Japan, Korea, Singapore and Vietnam) to report the community level toxic effects of such biomarkers under different climatic conditions. The second chapter emphasizes the importance of different fundamental/advanced biomolecular techniques (affinity- based chromatography) for the analysis of emerging contaminants in wastewater and the environment. This chapter highlights the importance of different binding agents such as antibodies, molecularly imprinted polymers, and chiral stationary phases with the bioanalytical derivatives like macrocyclic antibiotics, proteins and polysaccharide to have more insight on biological separation events. Chapters in this part provide an overview of various natural attenuation processes for CECs, with specific attention to aqueous photochemistry of pharmaceuticals, evaluated using laboratory-scale and on-site observations. Collectively, this part highlights natural attenuation for specific classes of pharmaceuticals and focused on the governing mechanisms -underlying such attenuation processes based on studies conducted worldwide. This part is also helpful in characterizing the various physico-chemical processes through which MPs enter into the river ecosystems and the fate of the plastic particles in aquatic environments. Moreover, the eco-toxicological effect of MPs towards the aquatic biota, their detection techniques and possible risk management have also been discussed to establish their abundance and to develop possible mitigation strategies to reduce human health hazard. The authors of the fifth chapter have summarized water quality analysis on a total of 1262 groundwater wells distributed all over the country (Sri Lanka) for the determination18 water quality parameters using multivariate statistical analysis techniques. Cluster analysis was used to classify the groundwater wells based on their quality. The purpose of the study is to increase the viability of water recourse management and protection in Sri Lanka by providing statistical analysis result for the entire country. This part also depicts the combined biological/physico-chemical processes such as natural biodegradation, phytoremediation, bioreactor, chemical reduction, adsorption, membrane filtration, ion exchange, electrochemical reduction etc. for the adsorptive removal of ClO_{4-} by iron nanoparticles, catalytic reactors etc. Among physical, chemical and biological ClO_{4-} removal methods, biological treatment is considered as the cheapest and eco-friendly technology. The final chapter of this part elucidates carcinogenic and human health properties of emerging contaminants such as nanoparticle and specific PPCPs. Arsenic, radon, hazardous waste, agricultural chemical, fluoride, etc. are discussed in detail focussing on their role in various types of cancer such as lungs, breast, kidney, bladder, liver etc. These kinds of critical discussions are needed to find economical ways to treat contaminated wastewater in order to reduce risk of cancer.

The eleven chapters in the second part reflect the multifaceted application of biological processes (bioreactor, anammox etc.), abiotic degradation (photo catalysis, ozonation, etc.), natural removal (constructed wetland), and granular sludge reactors as treatment techniques for the emerging contaminants. This diverse part provides an overview of critical concepts needed for evaluation of risk assessment methodologies, which in turn establishes the proper functioning of wastewater treatment methodologies for the effective CEC removal from wastewater. This part emphasizes various removal techniques utilizing photo catalysis, permeable reactive barrier (PRB), bioremediation, metal-organic framework, nanotechnology etc. and assesses the effectiveness of various wastewater treatment strategies. The first article describes case studies evaluating problems that occur during the installation of PRB in pilot-scale before scaling up. The authors have made a comparative study of different case studies, the filler material used, the type of construction used, the date of operational set-up and cost analysis of PRBs. The chapter concludes with an assessment of pros and cons of PRBs. The authors of the second chapter discuss the utility of microbes for chromium (Cr^{6+}) removal to understand the mechanism of Cr-tolerance and remediation through different microbes for an economic, easy and eco-friendly technique that can be used on the pilot and larger scale to ensure Cr6+ free water. Mixed culture bioreactors containing Cr^{6+}-tolerant microbes can be effectively used in treating Cr^{6+} contaminated wastewater as shown by the authors. The third chapter gives an exclusive review of the existing literature on PPCPs removal from water. Pros and cons of the reported PPCPs removal processes are reviewed and evaluated with respect to removal efficiency. The fourth chapter throws light on the advanced applications of nanotechnology in waste water treatment. The various types of nanomaterials such as carbon nanotubes, graphene based, nano composites, metal organic frameworks are discussed focusing on their structures and performances in removal of water contaminants. Bioremediation techniques for water purification and the toxicity of the nanomaterial after treatment are also discussed. This part include a study on the effectiveness and efficiency of bimetallic particles for dechlorination of chlorophenols (CPs) and the effect of degree of chlorination and iron oxide phases on CPs incorporation when using Ni/Fe bimetallic system. This study will surely advance our knowledge on CPs degradation mechanisms and advance removal techniques. An overview of antibiotics removal technologies from water and associated challenges of conventional and advanced treatment (sorption techniques, membrane processes, and ecological processes like constructed wetlands, integrated constructed wetland) are critically analyzed in this part. There is very little information available about the eco-toxicological effects of antibiotics on aquatic and terrestrial organisms. Therefore, the seventh chapter mainly deals with the health impact of antibiotics and antibiotic resistance (AR) and their removal techniques to estimate potential risks associated with the water treatment plants and effluent discharges to the environment. A critical assessment on the state-of-the art removal techniques, major pollutant detoxification pathways and their impact on groundwater flow regime are presented. This part highlights the basic science of the water quality treatment, occurrence/fate, and contaminant biodegradation along with the mobility and biological detoxification of pollutants. Chapter eight includes recent studies on beneficial chemicals/products and biogas generated from water hyacinth as well as its probabilities of success in commercialization. Chapter nine reviews literature on the uses of water hyacinth for phytoremediation as well as composting of water hyacinth and its application. Effective treatment of wastewater is essential for public health and sanitation, water reclamation, preventing environmental pollution and protecting water resources from contaminants getting introduced into surface water systems. The final chapter of this part has critically reviewed and summarized the currently available literature regarding the worldwide occurrence, fate and transport of perfluorooctane sulphonate (PFOS) in the environment. This study also discusses the various advanced removal processes like physical adsorption, membrane filtration and redox processes and other conventional removal technologies available for the sustainable remediation of PFOS. Literature gaps are identified and future directions are suggested accordingly. The future challenges of such different state-of-the-art techniques are discussed and provides readers with an overview of cost-benefits of the available technologies for effective in-situ treatment of CECs.

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Dr. Manish Kumar
Prof. Ryo Honda
Prof. Daniel D. Snow
Dr. Santanu Mukherjee

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Dr. Santanu Mukherjee obtained his Ph.D. from the University of Bonn and worked as a guest scientist in FZJ, Juelich. He was a visiting researcher in Savannah River Ecological Laboratory, USA. He is the recipient of multiple accolades such as EGU-Young Scientist Travel Award, ICAR international fellowship, GRIFA (Italian Pesticide Agency) grant, EU-COST action biochar grant, etc. His research interest includes the fate of emerging contaminants, the role of dissolved and particulate organic matter in deciding contaminants fate in the environment.

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Part I
Monitoring of Contaminants of Emerging Concern (CECs) in the Aquatic Environment

Chapter 1
Pharmaceuticals, Personal Care Products, and Artificial Sweeteners in Asian Groundwater: A Review

Keisuke Kuroda and Jun Kobayashi

1.1 Introduction

Pharmaceuticals and personal care products (PPCPs) are a group of contaminants of emerging concern (CECs) in the aquatic environment, which have gained global attention in the last 20 years because of their frequent occurrence and potentially adverse effects on human health and aquatic ecosystems (aus der Beek et al. 2016; Oaks et al. 2004; Schwarzenbach et al. 2006). The typical class of PPCPs found in the aquatic environment includes antibiotics, anti-inflammatories, analgesics, anticonvulsants, β-blockers, stimulants, contrast media, cosmetics, fragrances, insect repellents, and other diverse chemicals used for human and veterinary uses (Daughton and Ternes1999). Artificial sweeteners (ASs), which are sugar substitutes, have also been ubiquitously found in the water environment, owing to high human consumption and the ASs' high environmental persistence (Buerge et al. 2009; Kokotou et al. 2012). After consumption, both PPCPs and ASs are typically disposed of in wastewater via toilets, sinks, or bathrooms, and transported to wastewater treatment plants (WWTPs). There, because conventional WWTPs can only partly remove these compounds, their residues are discharged into receiving waters (Heberer 2002; Lange et al. 2012; Tambosi et al. 2010; Tran et al. 2018). There are also direct pathways of PPCPs and ASs into the water environment, such as sewage disposal, septic tank leakage, leakage from sewerage systems, livestock breeding, and fertilization (Sui et al. 2015).

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Asia, with a population of 4.6 billion as of 2020 (60% of the world population; https://population.un.org/wpp/), is a major consumer and producer of pharmaceuticals. For example, India and China were the two largest consumers of antibiotics in 2010 (Van Boeckel et al. 2014). In Asia, groundwater is an essential source of domestic water. In China, more than 400 of the country's 657 cities (61%) use groundwater as the major source of water supply (Liu and Zheng 2016). In India, around 80% of the rural population and 50% of the urban population use groundwater for domestic purposes (Chakraborti et al. 2011). Therefore, there is an urgent need to identify possible PPCP contamination in the groundwaters of various Asian regions.

The occurrence and fate of PPCPs and ASs in the world's groundwater have been summarized in a number of reviews (Lapworth et al. 2012; Postigo and Barceló 2015; Sui et al. 2015). These papers, however, rarely include studies on groundwater in Asia. This limited inclusion of Asian groundwater studies is partly because only a few Asian studies existed when the reviews were published. Since then, however, an increasing number of studies have been conducted on PPCPs and ASs in the groundwater of multiple Asian countries. Recently, a few country-specific review papers have been published on CECs, including PPCPs and ASs, in the groundwater of China (Dong et al. 2018) and India (Sackaria and Elango 2019). To date, however, there have been no reviews summarizing the results of studies investigating the occurrence of PPCPs and ASs in the groundwater of multiple Asian countries.

Hence, the objective of this chapter is to review the occurrence, sources, pathways, and fates of various pharmaceuticals, personal care products, and artificial sweeteners in groundwater across Asia. An overview of field studies from six Asian countries (China, India, Japan, Korea, Singapore, and Vietnam), as well as country-by-country analysis, is provided. The detection frequencies and concentrations of 13 representative PPCPs and ASs are compared among the studies. In addition, the main sources, pathways, and fates of the contaminants; the utility of PPCPs and ASs as anthropogenic markers or indicators of groundwater pollution; and the potential risk to humans and aquatic organisms; are summarized. Finally, future challenges are discussed from an Asian perspective.

1.2 Overview of PPCP and AS Studies on Asian Groundwater

Based on a database search using Scopus and Google Scholar, 22 scholarly publications were identified as suitable for analysis. The criteria for selection were: (i) the study involves detection of at least one PPCP or AS in Asian groundwater, (ii) the number of surveyed sites (wells) and samples are clearly specified, and (iii) the full text and its supplementary information are available in English. In addition, the results of the authors' recent investigation of groundwater in Tokyo were included. In total, 23 studies from six countries were summarized for review (Table 1.1). Most of the studies were conducted in China (10), followed by India (4), Japan (3), Vietnam (3), Singapore (2), and Korea (1).

The number of investigated groundwater sites per study ranged widely (from 3 to 50), as did the number of samples per study (3–148). We regarded spring water samples as groundwater samples because the properties of spring waters usually reflect those of nearby groundwaters. In 12 of the 23 studies (52%), information on the depth of groundwater wells or aquifers was obtained, with the latter ranging from very shallow (up to 2 m below ground level; hereafter, mbgl) to deep (up to 500 mbgl). In the other studies, where depth information was not provided, it appeared that most of the surveyed groundwater was shallow (<50 mbgl), based on the qualitative information in the papers.

The number of analyzed PPCPs and ASs also varied greatly, from 3 to 79 PPCPs and up to 7 ASs. Most of the studies employed targeted analysis with liquid chromatography-tandem mass spectrometry (LC-MS/MS) or gas chromatography-mass spectrometry (GC-MS). In the three studies, however, screening analysis of up to more than 1300 compounds (including 79 PPCPs) was employed, using GC-MS with automated identification and quantification database system (AIQS), and liquid chromatography time-of-flight mass spectrometry (LC-TOF-MS) (Duong et al. 2015; Kong et al. 2016; Li et al. 2016). As a general tendency, the number of detected PPCPs and ASs in the groundwater (up to 29 pharmaceuticals and 4 ASs) was less than the number of measured PPCPs and ASs in the respective studies. This would suggest that the groundwater was less contaminated, compared to surface waters, for example, because of the more direct pathways for the transport of contaminants in the case of the latter (e.g., direct discharge of wastewater effluent), as well as other factors such as differences in environmental fate and transport processes (e.g., sorption, volatilization, degradation, etc.), as evidenced by European (Loos et al. 2010) and US studies (Barnes et al. 2008; Focazio et al. 2008).

1.3 Country-by-Country Analysis

1.3.1 China

The occurrence of PPCPs and ASs in Chinese groundwater has been reported in 10 studies, the most among Asian countries. The geological scale of the groundwater studies in China varied widely, from the national (Li et al. 2015) and regional scale (Kong et al. 2016; Li et al. 2016) to watershed (Gan et al. 2013; Jiang et al. 2019; Xiang et al. 2018; Yang et al. 2018) and local scale (Peng et al. 2014; Tong et al. 2014; Yao et al. 2018). Dong et al. (2018) summarized the occurrence of a wide range of CECs, including PPCPs and ASs, in groundwater in China. This chapter summarizes seven further papers that were not discussed in Dong et al. (2018).

Table 1.1 Summary of studies on PPCPs and ASs in Asian groundwater

Country	Location	Number of site/sample	Depth[a] (mbgl)	Compounds analyzed	Compounds detected	LOD[b]/LOQ[c] (ng/L)	Maximum conc. (ng/L)	References
China	Tianjin	5/15	n.a. [d]	7 ASs	4 ASs (acesulfame, cyclamate, saccharin, sucralose)	0.1–2.3 (LOD), 0.4–7.5 (LOQ)	Cyclamate (100)	Gan et al. (2013)
	Guangzhou	33/132	<10	13 antibiotics, 15 other pharmaceuticals, 8 personal care products (PCPs), 1 industrial chemical	4 antibiotics, 11 other pharmaceuticals, 7 PCPs, 1 industrial chemical	0.1–6 (LOQ)	Salicylic acid (2014.7)	Peng et al. (2014)
	Shahu County, central China	27/44	15–100	19 antibiotics	19 antibiotics	0.05–5.3 (LOQ)	Erythromycin dehydrate (377.8)	Tong et al. (2014)
	12 cities from northern China	12/12	5–70	11 other pharmaceuticals, 5 endocrine-disrupting chemicals (EDCs)	4 other pharmaceuticals, 5 EDCs	0.01–1 (LOD), 0.004–5.5 (LOQ)	Sulpiride (60.1)	Li et al. (2015)

(continued)

Table 1.1 (continued)

Country	Location	Number of site/sample	Depth[a] (mbgl)	Compounds analyzed	Compounds detected	LOD[b]/LOQ[c] (ng/L)	Maximum conc. (ng/L)	References
	Dalian rural areas	13/13	2.5–100	21 antibiotics, 58 other pharmaceuticals as well as more than 1221 organic chemicals	1 antibiotic (sulfamethoxazole), 3 other pharmaceuticals, 1 PCP (L-menthol)	4–20 (LOD)	Nicotine (689)	Li et al. (2016)
	Beijing and Tianjin	27/27	n.a.	21 antibiotics, 58 other pharmaceuticals as well as more than 1221 organic chemicals	1 antibiotic (oleandomycin), 3 other pharmaceuticals, 3 PCPs	4–200 (LOQ)	2-phenoxy-ethanol (1328)	Kong et al. (2016)
	Shahu County, central China	39/99	10, 25 or 50	14 antibiotics	14 antibiotics	0.01–2.16 (LOD)	Clorotetracycline (59.6)	Yao et al. (2017)
	Dongjiang River Basin	11/33	2–13	48 antibiotics, 6 X-ray contrast media, 24 other pharmaceuticals, 8 PCPs, 5 ASs, 7 other organic chemicals	6 antibiotics, 2 X-ray contrast media, 10 other pharmaceuticals, 6 PCPs, 4 ASs, 5 other organic chemicals	0.03–153 (LOD), 0.01–45.8 (LOQ)	Acesulfame (4580)	Yang et al. (2018)

(continued)

Table 1.1 (continued)

Country	Location	Number of site/sample	Depth[a] (mbgl)	Compounds analyzed	Compounds detected	LOD[b]/LOQ[c] (ng/L)	Maximum conc. (ng/L)	References
China	Shanghai	3/3	n.a.	13 psychiatric pharmaceuticals, 2 metabolites	9 psychiatric pharmaceuticals	0.05–1.06 (LOD), 0.17–3.54 (LOQ)	Lorazepam (46.83)	Xiang et al. (2018)
	Changzhou, Yangtze River Delta	5/5	n.a.	27 antibiotics, 16 other pharmaceuticals, 1 PCP	16 antibiotics, 13 other pharmaceuticals, 1 PCP	0.02–14.75 (LOD), 0.09–58.99 (LOQ)	Caffeine (503.76)	Jiang et al. (2019)
India	Patancheru, Telangana	5/5	n.a.	7 antibiotics, 5 other pharmaceuticals	5 antibiotics, 5 other pharmaceuticals	10–50 (LOQ)	Cetirizine (28,000)	Fick et al. (2009)
	Patancheru, Telangana	15/26	n.a.	3 fluoroquinolone antibiotics	3 fluoroquinolone antibiotics	10 (LOQ)	Ciprofloxacin (770)	Rutgersson et al. (2014)
	Varanasi, Uttar Pradesh	26/26	10–200	3 antibiotics, 11 other pharmaceuticals, 1 AS, 1 industrial chemical, 21 pesticides and metabolites, 3 per- and polyfluoroalkyl substances (PFASs)	3 antibiotics, 10 other pharmaceuticals, 1 AS, 11 pesticides and metabolites, 3 PFASs	0.1–100 (LOD)	Sulfamethoxazole (120)	Lapworth et al. (2018)

(continued)

Table 1.1 (continued)

Country	Location	Number of site/sample	Depth[a] (mbgl)	Compounds analyzed	Compounds detected	LOD[b]/LOQ[c] (ng/L)	Maximum conc. (ng/L)	References
	Ganges River Basin	14/14	n.a.[4]	2 antibiotics, 10 other pharmaceuticals, 3 PCPs, 5 ASs	2 antibiotics, 8 pharmaceuticals, 3 PCP, 3 ASs	0.06–15 (LOD)	Caffeine (262)	Sharma et al. (2019)
Japan	Central Tokyo	15/15	10–33	11 other pharmaceuticals, 2 PCPs	7 other pharmaceuticals, 2 PCPs	0.1–12 (LOQ)	Crotamiton (592)	Nakada et al. (2008)
	Central Tokyo	50/50	10–500	5 other pharmaceuticals, 1 PCP	5 other pharmaceuticals, 1 PCP	0.10–21 (LOQ)	Crotamiton (1400)	Kuroda et al. (2012)
	Central Tokyo	14/14	10–30	5 antibiotics, 10 other pharmaceuticals, 2 PCPs	5 antibiotics, 10 other pharmaceuticals	0.28–3.47 (LOQ)	Ibuprofen (1006)	This study
Korea	Buyeo-gun	30/30	10–15	17 antibiotics, 15 other pharmaceuticals, 1 PCP, 5 ASs, 6 pesticides	14 antibiotics, 11 other pharmaceuticals, 1 PCP, 1 pesticide, 4 ASs	0.03–4 (LOD)	Acesulfame (1330)	Lee et al. (2019)

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