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Human health risk assessment of pharmaceuticals in treated wastewater reused for non-potable applications in Sharjah, United Arab Emirates

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ABSTRACT

Pharmaceuticals and personal care products are an integral part of societal health yet their presence in various environmental compartments, including treated wastewaters, has sparked concerns towards possible human and ecological health effects. The current study aims to characterize human health risks posed by ten pharmaceuticals quantified in wastewater treatment plant effluents where water is reused mainly for landscape irrigation. Receptors were identified as children playing in green areas, adult landscape workers, and adult users of athletic and golf courses irrigated by treated wastewater. The human health risk assessment model exhibited safe exposure (RQ $<$ 1) to all pharmaceuticals for all receptors through both dermal and ingestion exposure pathways. RQs were highest for the landscape worker followed by children playing in green areas and then adult using the athletic fields. RQs were highest to lowest in the following order of pharmaceuticals: acetaminophen, metoprolol, ciprofloxacin, erythromycin, ofloxacin, sulfadiazine, sulfamethoxazole, sulfapyridine, risperidone, and sulfamethazine. Such risk assessment findings aid in supporting decisions to optimize wastewater treatment and reuse strategies, as well as safeguard public and environmental health.

1. Introduction

Wastewater recycling and reuse has attracted remarkable attention during recent decades as an alternative source of water. Communities have been using reclaimed water to irrigate landscapes, forests, and agricultural fields; provide water-consuming industries with an alternative to freshwater; and to supplement stream flows and groundwater aquifers. Although most water reuse initiatives have been developed to meet non-potable water demands, a number of projects use recycled water indirectly for potable purposes (USEPA, 2017; Maiolo and Pantusa, 2017; Angelakis and Snyder, 2015; PUB, 2015; Mudgal et al., 2015; Angelakis and Gikas, 2014; Tchobanoglous et al., 2011). Adequately treated recycled water, with a quality appropriate for the intended use, can satisfy most water demands. In reuse applications where there is a greater potential of human exposure to the recycled water, more treatment is certainly required to safeguard public health.

Wastewater reuse applications play an integral role in meeting water demands in the United Arab Emirates (UAE). Wastewater reuse for landscape irrigation in particular is widely practiced in the City of Sharjah and other cities in the UAE as a water management strategy to alleviate the country's water scarcity and to promote environmental sustainability and protection (Gulf News, 2017; FAO, 2015; World Bank., 2011; ACWUA, 2010). In recent years, interest in the public health ramifications of trace levels of pharmaceuticals and personal care products (PPCPs) in reused wastewater has been increasing worldwide as most municipal reclamation plants are not specifically designed to deal with the trace levels of such contaminants of emerging concern (CECs). Further, many of these compounds pass through conventional treatment systems without efficient removal (Ebele et al., 2017; Yang et al., 2017; Sui et al., 2015; Petrie et al., 2015; Luo et al., 2014; Guerra et al., 2014) resulting in trace concentrations of PPCPs in various environmental matrices, including treated wastewater effluents. Various potential environmental effects of PPCPs such as chronic toxicity (significant decrease in fecundity of species under study such as cladoceran *Daphnia magna* and fish), endocrine disruption in aquatic wildlife, and development of bacterial pathogen resistance (Damasceno de Oliveira et al., 2016; Overturf et al., 2015; Petrie et al., 2015; Marti et al., 2014) have been revealed. Substantial work has been

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conducted to evaluate potential health effects of PPCPs present in drinking water sources and remaining in finished drinking water (Arlos et al., 2015; Sun et al., 2015; Simazaki et al., 2015; Gaffney et al., 2015; Wen et al., 2014) yet little information on potential health effects associated with these chemicals in the context of non-potable reuse applications is available.

To best assess human health risks and frame safe target concentrations for water and wastewater treatment, established risk assessment methods accepted by regulatory authorities such as United States Environmental Protection Agency (USEPA) can be used (USEPA, 2010, 2004, 1991, 1989). Several risk assessment studies have been conducted to assess potential human health risks from water reuse for potable purposes (Lin et al., 2016; Bruce et al., 2010; Kumar et al., 2010; Schwab et al., 2005; Webb et al., 2003), through ingestion of crops irrigated with treated wastewater (Koopaei and Abdollahi, 2017; Ebele et al., 2017; Prosser and Sibley, 2015; Roccaro and Vagliasindi, 2014; Kumar et al., 2010) and through consumption of fish from surface waters receiving treated wastewaters (Kumar et al., 2010). Yet, human health risk assessment of wastewater reuse for non-potable uses is very limited (Kennedy et al., 2012).

The current study focuses on conducting a quantitative human health risk assessment for a group of pharmaceuticals identified and quantified in Sharjah Wastewater Treatment Plant (SWWTP) effluents where the treated wastewater is reused for non-potable purposes, mainly landscape irrigation. Findings will be of significance towards supporting decisions to optimize wastewater treatment and reuse strategies, as well as safeguard public and environmental health. The study will also serve as a significant baseline research in view of the very limited availability of published research on occurrence of pharmaceuticals in UAE environmental systems as well as on associated health risk assessments.

2. Hazard identification

In the current study, the hazards have been considered to be a group of pharmaceuticals identified and quantified in SWWTP effluents during the period of January to November 2017 (Table 1) where the treated wastewater is reused for non-potable purposes. Risk assessment has been subsequently performed on minimum, maximum, and average pharmaceutical concentrations reported in monthly composite effluent wastewater samples collected from the SWWTP under study. Composite samples from the effluent wastewater of SWWTP have been collected properly into pre-rinsed dry amber glass bottles. Wastewater samples were collected every 90 min then composited to accommodate for variations in wastewater flows and pharmaceutical concentrations at varying sampling episodes; thus, providing samples with better representation. All collected samples have been properly sealed and transferred to the lab by icebox. In the laboratory, the samples have been filtered under vacuum through 0.7 μm glass fiber filters and kept at 4 °C in the dark for a maximum period of 1 week until extraction. Target pharmaceuticals have been selected based on their abundant usage and analyzed by the authors using Waters Acquity® UPLC H-Class-Xevo TQD (Triple Quadrupole Mass Spectrometer) system (MA, USA) equipped with electrospray ionization (ESI). Chromatographic separation of the target compounds has been achieved on Acquity® BEH C18 column (1.7 μm, 2.1 mm × 150 mm) using gradient elution of two mobile phases: methanol and 0.2% formic acid in 5 mM ammonium formate. The flow rate has been set at 0.2 mL/min and the injection volume at 10 μL. All of the obtained validation parameters of the method satisfied the requirements and guidelines of analytical method validation as correlation coefficient values in the linear calibration plot for each target compound exceeded 0.99 and the recovery percentages of the investigated pharmaceuticals were > 84%. Limit of detection (LOD) varied between 0.1 and 1.5 ng/L and limit of quantification (LOQ) was 0.3–5 ng/L for all analytes. The precision of the method was found to be in the ranges of 2.2% to 7.7% and 2.2% to 8.6% for inter and intra-day analysis, respectively (Semreen et al., submitted for publication).

Table 1
Concentrations of pharmaceuticals in SWWTP influents and effluents (Jan–Nov 2017).

Analyte	Concentration (ng/L)	
	Influent range (average)	Effluent range (average)
Sulfapyridine	86–417 (251.7)	89–111 (99.9)
Sulfamethazine	11–36 (23.7)	7–15 (11.0)
Sulfadiazine	554–886 (720.2)	268–599 (433.4)
Sulfamethoxazole	96–228 (161.8)	69–81 (75.1)
Ciprofloxacin	697–1028 (862.7)	378–709 (543.4)
Ofloxacin	680–1012 (845.9)	345–676 (510.8)
Erythromycin	619–951 (785.2)	375–707 (541.2)
Acetaminophen	143,905–146,596 (145,250.3)	3890–6581 (5235.3)
Metoprolol	75–109 (92.1)	46–79 (62.5)
Risperidone	189–300 (244.8)	11–15 (13.2)

3. Toxicity assessment

The toxicity assessment for this study is based on literature review and findings from recent studies on the toxicological relevance of PPCPs in which toxicological benchmarks for such CECs were established. Potential relevant sources of information included drug databases (www.drugbank.ca; www.mayoclinic.org; www.drugs.com), National Library of Medicine PubMed database, and documents prepared by WateReuse Research Foundation, USEPA, World Health Organization (WHO), and others. Typically, for non-carcinogenic end points, threshold doses for toxicological effects are identified from human and/or animal studies which identify no observed adverse effect levels (NOAELs) and lowest observed adverse effect levels (LOAELs) for developmental, systemic, reproductive, and other toxicity endpoints (Bruce et al., 2010). For risk assessment of pharmaceuticals, minimal therapeutic doses from approved drug labels may also be used in toxicological assessment. NOAELs, LOAELs, and therapeutic doses are later divided by uncertainty factors (UF) to account for uncertainties in the data set or accommodate for potentially sensitive populations (Kennedy et al., 2012; WHO, 2012; Bull et al., 2011; Bruce et al., 2010; DWI, 2007).

In this study, the toxicological benchmarks are expressed as acceptable daily intakes (ADI) and represent the daily intakes of pharmaceuticals that are unlikely to result in adverse health effects to humans, including sensitive population subgroups. The ADIs were identified for the pharmaceutical compounds under study based on a hierarchy developed by Snyder et al. (2010) and adopted in similar studies of risk assessment of wastewater reuse for non-potable uses (Kennedy et al., 2012). The hierarchy proceeds according to the following manner; for pharmaceutical target compounds, the lowest value is selected among the values calculated as follows:

(i) Therapeutic dose in mg/kg-day (based on prescription range of doses and age groups) divided by a default UF = 3000. If the compound is either a non-genotoxic carcinogen or an endocrine disrupting chemical (EDC), the dose should be divided by and additional UF of 10.
(ii) NOAEL divided by a default UF = 1000 or LOAEL divided by a default UF = 3000. An additional UF = 10 is considered if the compound is either a non-genotoxic carcinogen or an endocrine disrupting chemical (EDC).
(iii) If the compound is a genotoxic carcinogen and tumor incidence data are available, develop a slope factor and establish a comparison value assuming a safe minimum cancer risk of 1:1,000,000.
(iv) If the compound is a genotoxic carcinogen and no tumor incidence data are available, use the lower of the virtually safe dose derived