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Assessment of drugs and personal care products biomarkers in the influent and effluent of two wastewater treatment plants in Ho Chi Minh City, Vietnam

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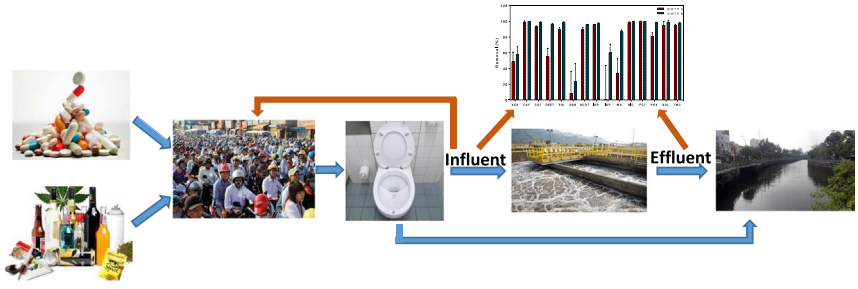
HIGHLIGHTS

- The occurrence and consumption of 19 drugs and personal care products biomarkers were assessed in wastewater influent.
- Paracetamol and caffeine consumption were highest at 320 g/d/1000 pp and 300 g/d/1000 pp, respectively.
- Removal and release of DPCPBs from wastewater were also taken into account.
- An estimated 167 kg per day of the studied DPCPBs are released into environmental surface waters.

GRAPHICAL ABSTRACT



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ABSTRACT

Wastewater samples were collected at the influent and effluent of two wastewater treatment plants (WWTPs) in Ho Chi Minh City, Vietnam and then pooled to daily samples over multiple days using 6 hourly grab samples. The aim was to provide a first assessment of the occurrence, consumption, removal and release of a range of organic chemicals including pharmaceuticals and personal care products (PPCPs), illicit drugs, an artificial sweetener, tobacco and its metabolites and alcohol biomarkers (referred to here as DPCPBs). Nineteen DPCPBs were detected via direct measurement of filtered wastewater on LC-MS/MS with a concentration range of 0.05–38 μg/L. Caffeine and paracetamol were the most prominent compounds detected in the influent, while acesulfame was found at the highest concentration in the effluent of both WWTPs. Mean concentrations of metabolites of tobacco (nicotine: 7.6 μg/L, cotinine: 1.4 μg/L and hydroxycotinine: 1.7 μg/L) and alcohol (ethyl sulphate: 3.3 μg/L) were lower than those of European countries. Consumption rates based on daily mass loads and catchment population data obtained from the WWTPs were <10 g/d/1000 pp for the majority of selected PPCPs, except for caffeine (300 g/d/1000 pp) and paracetamol (320 g/d/1000 pp). Consumption rates for codeine and methamphetamine were 0.05 g/d/1000 pp and 0.17 g/d/1000 pp, respectively. Consistently across the two WWTPs most of the chemicals (10) showed >80% apparent removal rate from the wastewater, three chemicals showed apparent removal efficiency of approximately 50%; and the removal efficiency could not be assessed for 5 compounds due to their low concentrations in the influent. Based on the fraction of treated and untreated wastewater (10:90) that is released into the receiving environment we estimated a total discharge of approximately 170 kg per day of DPCPBs in Ho Chi Minh City.

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1. Introduction

Vietnam is growing both in terms of population and economy. With this growth, the use of pharmaceuticals and personal care products (PPCPs), illicit drugs, tobacco and alcohol has also been rapidly increasing in recent years (McKetin et al., 2008; Tran et al., 2013). Consequently, it is expected that the levels of these organic chemicals have also increased in wastewater in Vietnam, although data on presence and levels of organic chemicals in Vietnamese domestic wastewater treatment plants (WWTPs) is very scarce. Only a few studies so far have reported concentrations of PPCPs in wastewater and hospital effluent (Tran et al., 2014; Watanabe et al., 2016). Therefore, information about the loading of organic chemicals in and out of WWTPs provides important evidence towards better environmental management.

Systematic sampling and analysis of wastewater has rapidly evolved in the past decade to provide spatial and temporal information on presence and consumption of organic chemicals in different catchments (EMCDDA, 2016). A wide range of organic chemicals such as pharmaceuticals and personal care products (PPCPs), illicit drugs (IDs), tobacco and alcohol (referred to as drugs and personal care products biomarkers (DPCPBs)), have been measured in WWTP influent and used to estimate their consumption worldwide (Castiglioni et al., 2015; Hall et al., 2012; Kim et al., 2015; Lai et al., 2013a,c; Prichard et al., 2012; Ryu et al., 2016). Many relatively persistent DPCPBs have been found in WWTPs effluent showing that WWTPs are important contributors of these chemicals to the environment (Petrović et al., 2003). For this reason, WWTP effluent should be monitored to obtain a better understanding of WWTPs performance for organic chemical removal and inform risk assessments for the receiving environment.

Ho Chi Minh City is the largest metropolitan area in Vietnam with a population of approximately 9 million, generating a volume of domestic wastewater of around 2 million m^3 per day (DONRE, 2013; Nguyen, 2010). Nevertheless, the wastewater treatment capacity in Ho Chi Minh City is very limited with <10% of the wastewater being collected and treated in WWTPs whereas the remaining wastewater released to the surface water is untreated (Le et al., 2013). The few operational WWTPs are among the first in Vietnam (Le et al., 2013), hence they provide an opportunity to apply wastewater-based epidemiology and assess chemical use in the population of Ho Chi Minh City. Furthermore, data on concentrations of DPCPBs in influent provide an estimate of their release into the environment from untreated wastewater and in combination with data from effluents can be used to provide an estimate of total release from treated and untreated wastewater into the environment.

The aims of this study were to evaluate for the first time the presence and removal efficiency of a wide range of DPCPBs in two WWTPs in Ho Chi Minh City, Vietnam. Mass loads of DPCPBs in the influent and effluent were used to estimate human consumption rates of some compounds, and the environmental discharge into surface waters of the city.

2. Materials and methods

2.1. Chemicals and reagents

Analytical grade acetic acid was purchased from Sigma Aldrich (Castle Hill, Australia). Analytical grade hydrochloric acid 32% was purchased from Univar (Ingleburn, Australia). Liquid chromatography grade methanol was purchased from Merck (Darmstadt, Germany). Water was purified through a MilliQ system (Millipore, 0.22 μm filtered, 18.2 mΩ cm^{-1}). Native and labelled analytical standards of target compounds were obtained from various suppliers. Calibration standards were prepared in MilliQ water acidified to pH 2 with hydrochloric acid.

2.2. Description of the WWTPs

Wastewater samples were collected from two WWTPs in Ho Chi Minh City, referred to as WWTP A and WWTP B. The treatment capacity of WWTP A is 140,000 m^3/day serving about 425,000 inhabitants, using conventional activated sludge treatment. The treatment capacity of WWTP B is 26,000 m^3/day serving 120,000 inhabitants, with aerated ponds technology. A summary description of the two WWTPs is provided in Table S2. A portion of wastewater, called black wastewater, estimated to be approximately 92% (Le et al., 2013), goes through septic tanks prior to entering the centralized sewage system linked to the WWTPs. The septic tanks retain black wastewater until the wastewater overflows into the sewer system thus increasing the retention of the wastewater in the sewer system. Approximately 8% grey domestic wastewater generated from household except for the wastewater from the toilet was released directly to the sewage system (Le et al., 2013).

2.3. Sample collection and preservation

Due to the unavailability of autosamplers, the sampling campaign relied on collection of multiple grab samples. Wastewater samples ($n = 80$) were collected from the influent and effluent of the two WWTPs to estimate the chemical loads coming to the WWTP and the apparent removal rates. Approximately 500 mL of sample was collected every 6 h over a 7-day period for WWTP A (8–14 December 2015) and a 3-day period for WWTP B (20–22 December 2015). All samples were acidified immediately after collection using 1 M hydrochloric acid (pH 2). Samples were first stored at 4 °C for one week at Ho Chi Minh University of Science. Samples were frozen at −20 °C before being shipped on ice to Australia by air and then kept at −20 °C at the Queensland Alliance for Environmental Health Sciences (QAEHS), The University of Queensland until analysis.

2.4. Sample preparation

Prior to analysis, samples were thawed and pooled to obtain daily composite samples by mixing four samples that were collected on the same day (50 mL × 4). In total, 20 pooled samples of influent and effluent were made up from 80 initial wastewater samples. The sampling and pooling of samples in this study is equivalent to a time-proportional sampling mode with large interval. Such large interval may affect the accuracy of the estimated chemical loads, which is a limitation of this study.

Subsequently, 1 mL of each pooled sample was filtered by 0.45 μm RC syringe filter (Phenomenex, Lane Cove, Australia) and transferred into a 1.5 mL glass amber vial. The samples were spiked with labelled internal standards (Table S1) prior to being injected onto LC-MS/MS.

2.5. LC-MS/MS analysis

The samples were screened for 110 commonly used illicit drugs, PPCPs, tobacco and alcohol metabolites (Table S1). Of these, nineteen compounds were detected and quantified in the samples (Table 1).

PPCPs were analyzed by a LC-MS/MS method (O'Brien et al., 2017) using a Sciex 6500QTRAP mass spectrometer (Sciex, Concord, Ontario, Canada) equipped with an electrospray (ESI) (TurboV) interface coupled to a Shimadzu Nexera HPLC system (Shimadzu Corp., Kyoto, Japan). Separation was achieved on a Kinetex Biphenyl column (2.6 μm, 100 Å, LC Column 50 mm × 2.1 mm, Phenomenex) with a mobile phase gradient of 1 to 95% methanol with 0.1% acetic acid. Illicit drug residue analysis was conducted as outlined by Lai et al. (2013b) and tobacco and alcohol metabolite analysis was conducted as outlined by Banks et al. (2017). These methods were performed on a Sciex 5500QTRAP mass spectrometer with an ESI interface (Sciex, Concord, Ontario, Canada) coupled to Shimadzu Nexera HPLC systems (Shimadzu