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Occurrence and fate of illicit drugs and pharmaceuticals in wastewater from two wastewater treatment plants in Costa Rica

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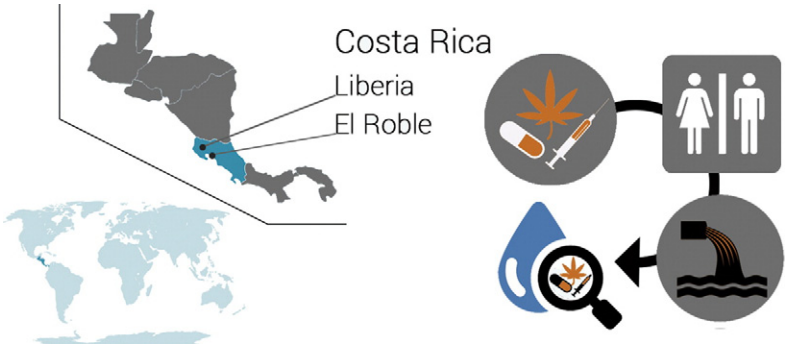
HIGHLIGHTS

- Wastewater-based epidemiology first application in Central America
- Key parameters assessment to apply best practice protocol
- LC-HRMS analysis allowed target and suspect screening.
- Cocaine and cannabis were found to be the most consumed drugs in Costa Rica.
- Residues at low concentration were detected in surface water samples.

GRAPHICAL ABSTRACT



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ABSTRACT

Chemical analysis of raw wastewater in order to assess the presence of biological markers entering a wastewater treatment plant can provide objective information about the health and lifestyle of the population connected to the sewer system. This work was performed in a tropical country of Central America, Costa Rica, with the aim of extending this knowledge to new world regions. This work is the first to report wastewater-based epidemiological data on the use of illicit drugs in this region of the world. Composite wastewater samples from the influents of two different wastewater treatment facilities and surface water samples from surrounding areas were collected applying the best practice protocol and analysed to investigate the occurrence and fate of selected illicit drugs of abuse and pharmaceuticals. Results showed the presence of chemical indicators of the classic drugs cocaine and cannabis at high concentration levels, besides the moderate presence of the opiates codeine and morphine. Neither the worldwide commonly used psychoactive substances of abuse such as synthetic phenethylamines, nor pharmaceuticals from the family of benzodiazepines were detected, demonstrating the spatial differences in drug use among different world regions. In addition, effluent wastewater samples were analysed and compared to influent concentrations in order to evaluate the decrease in concentration of the targeted analytes through two treatment technologies. As a final step, a wide-scope qualitative screening, including hundreds of suspect compounds, was applied in order to have a better knowledge on the presence of pharmaceuticals in waters and to assess the potential impact of the treated wastewater into the receiving aquatic ecosystems.

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1. Introduction

Chemical analysis of wastewater influents entering wastewater treatment plants (WWTPs) can provide valuable information on the health and lifestyle of the community connected to the sewer system (Daughton, 2001). In recent years several studies have been published showing the variety of licit and illicit substances which can be found in wastewater (Causanilles et al., 2016; Ort et al., 2014; Thomas et al., 2012). Loads of illicit drugs and pharmaceuticals are transported to municipal WWTPs where their concentrations can be determined by sophisticated analytical methodologies, and the results used to estimate drug use by the population. This approach, known as wastewater-based epidemiology, has allowed comparative studies between different urban centres around the world. Since the first European collaborative study in 2012 (Thomas et al., 2012), many other researchers have published their results, and the interest has been expanded to other world regions including America and Asia (Bijlsma et al., 2016; Devault et al., 2016; Du et al., 2015; Gatidou et al., 2016; Kim et al., 2015; Klupczynska et al., 2016; Lai et al., 2016). This type of studies that focus on the determination of drug residues in the raw sewage is now recognised as a complimentary tool in the assessment of drug prevalence (Amundsen and Reid, 2014; van Wel et al., 2016).

Besides this epidemiology point of view, there are also environmental implications, because the composition of raw sewage gives information that helps to decide what is the most appropriate treatment to remove or minimise those pollutants and prevent their discharge to the environment (Mara, 2003). Earlier studies in Europe (Bijlsma et al., 2012; Camacho-Muñoz et al., 2009) have shown this potential environmental threat that results from the fact that WWTPs are not specifically designed for removing illicit drug chemical indicators. Actually, some of these, such as 3,4-methylenedioxymethamphetamine (MDMA), are not removed at all by the treatment thus resulting in actual discharges into surface waters. In developing countries it is important that the treatment of municipal wastewater is efficient as much as sustainable, simple in its maintenance, and low in energy and chemicals usage (Mara, 2003). These considerations should be taken into account in order to implement the most appropriate treatment for the specific circumstances of the wastewater to be treated (which are not necessarily the same as in industrialized areas). Treatment processes can vary from wastewater stabilization ponds (WSP), where the wastewater flows at very low flow rate to enable removal by natural processes facilitated by sunlight and high temperatures during a determined residence time, to more technologically advanced systems, more appropriate when the land area required in natural systems is not available (Mara, 2003).

The study presented in this paper was performed at two locations of Costa Rica, in Central America. The goals were to investigate the presence of 15 selected chemical residues of illicit substances and pharmaceuticals in raw wastewater entering two WWTPs with different treatment technologies. The analytical information obtained was used in an attempt to apply the wastewater-based epidemiology approach for the first time to Costa Rican communities as well as to relate the loads observed to consumption by the population connected to the sewer system. Given the different treatment processes employed in the two WWTPs included in the study, their potential to reduce loads of illicit drugs and pharmaceuticals into the aquatic environment was assessed as well. Finally, since very few data are available for the occurrence of pharmaceuticals in the aquatic environment of Central America and in order to evaluate the potential impact of the wastewaters, a wide-scope qualitative screening was applied to influent and effluent wastewater as well as to surface water down and upstream from the effluent discharge point.

2. Materials and methods

2.1. Sampling sites

Samples were collected in two areas of Costa Rica, at two different WWTPs (see map in Fig. 1). The first WWTP sampled is located in the northwest of the country and serves the city of Liberia (capital of Guanacaste province). The treatment process consists of 4 WSP, connected in two series. Each pond measures 265 m long, 60 m wide and 2 m deep, containing up to 30,000 m^3 of water. The average influent flow rate is 2,680 m^3 per day and the hydraulic retention time varies from 24.3 days for the primary ponds to 31.5 days for the secondary ponds, being the total retention time of 55.8 days (Abarca Garbanzo, 2000). See a diagram of the system in Fig. S.1. The second WWTP sampled is located in the central west part of the country in the neighbourhood of El Roble and serves the city of Puntarenas. The process at this WWTP consists of primary settling and secondary treatment with Integrated Fixed-film Activated Sludge (IFAS). The influent wastewater is pumped to the entrance where it is separated into two parallel streams. The average influent flow rate is 6,650 m^3 per day. The two WWTPs were selected for the study based on the difference in treatment technologies and to further investigate the presence of pharmaceuticals and illicit drug residues found in a preliminary collection of grab samples.

In addition, surface waters were collected at two rivers nearby: the Liberia and Tárcoles rivers. The Liberia river flows through the city of Liberia, receiving the effluent discharged from the WSPs. It is a small tributary that discharges into the Tempisque river on its way to the Gulf of Nicoya. The river Tempisque is 144 km long. The Tárcoles river originates on the southern slopes of the *Cordillera Central* volcanic range and flows in a south-westerly direction to the Gulf of Nicoya. The river is 111 km long and its watershed encompasses around 50% of the country's population based at the central valley. See Fig. 1 for information on the sampling locations.

2.2. Sewer system characterization

The two sewer systems were characterized by means of a standardized questionnaire developed by Ort and colleagues (Castiglioni et al., 2012). The information describes relevant catchment properties, such as number of inhabitants connected to the sewer system and its basis of the estimation, the type of sewer drains, whether exfiltration is expected or not, influent flow control and flow profile or variations. Wastewater chemical properties were also measured: pH, biochemical and chemical oxygen demand (BOD_5 and COD) and organic nitrogen content (Nitrogen-Kjeldahl) (Andreottola et al., 1994). A summary is presented in Table S.1. The collection of this meta-data is essential for the correct interpretation of data obtained and their normalization for further comparison.

2.3. Sample collection

Different types of samples were collected in order to address the different goals (see Table S.2 for details):

A. 24-h composite influent wastewater samples at both WWTPs to relate the presence of residues of illegal drugs and pharmaceuticals to their consumption by the community included in the catchment area.
B. Grab effluent wastewater samples to describe the efficiency of both treatment systems in removing those residues.
C. Grab river surface water samples to investigate whether those residues can be observed in the aquatic environment.
D. Pooled surface water samples at the different WSP of the system at Liberia to gain more knowledge on the fate of chemicals along this natural treatment process.