



Occurrence patterns of pharmaceutical residues in wastewater, surface water and groundwater of Nairobi and Kisumu city, Kenya



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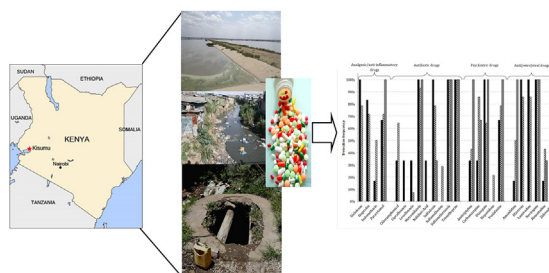
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HIGHLIGHTS

- 24 pharmaceuticals detected in Kenyan wastewater, river water and groundwater.
- Highest detection frequencies within the classes of antibiotics and anti(-retro)virals.
- Wastewater stabilization ponds remove pharmaceuticals with varying efficiency.
- River water concentrations similar as in wastewater; loads up to 180 kg d^{-1} .
- $>1 \mu\text{g L}^{-1}$ of the antiretroviral nevirapine in shallow wells used as drinking water.

GRAPHICAL ABSTRACT



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ABSTRACT

Emerging organic contaminants have not received a lot of attention in developing countries, particularly Africa, although problems regarding water quantity and quality are often even more severe than in more developed regions. This study presents general water quality parameters as well as unique data on concentrations and loads of 24 pharmaceuticals including antibiotic, anti(RETRO)viral, analgesic, anti-inflammatory and psychiatric drugs in three wastewater treatment plants, three rivers and three groundwater wells in Nairobi and Kisumu. This allowed studying removal efficiencies in wastewater treatment, identifying important sources of pharmaceutical pollution and distinguishing dilution effects from natural attenuation in rivers. In general, antiretrovirals and antibiotics, being important in the treatment of common African diseases such as HIV and malaria, were in all matrices more prevalent as compared to the Western world. Wastewater stabilization ponds removed pharmaceuticals with an efficiency between 11 and 99%. Despite this large range, a different removal is observed for a number of compounds, as compared to more conventional activated sludge systems. Total concentrations in river water (up to $320 \mu\text{g L}^{-1}$) were similar or exceeded concentrations in untreated wastewater, with domestic discharges from slums, wastewater treatment plant effluent and waste dumpsites identified as important sources. In shallow wells situated next to pit latrines and used for drinking water, the recalcitrant antiretroviral nevirapine was measured at concentrations as high as $1-2 \mu\text{g L}^{-1}$. Overall, distinct pharmaceutical contamination patterns as compared to the Western world can be concluded, which might be a trigger for further research in developing regions.

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1. Introduction

In the last three decades, there has been an increasing concern about the environmental occurrence and impact of pharmaceuticals, being a large and diverse group of emerging organic micro-pollutants. Pharmaceuticals are often poorly removed by conventional wastewater treatment techniques, and behave as (pseudo-)persistent contaminants in the environment (de Graaff et al., 2011; Fernández et al., 2014; Mceneff et al., 2014; Rivera-Utrilla et al., 2013; Verlicchi and Zambello, 2014). Consequently, they have been ubiquitously detected in (treated) wastewater (Deblonde et al., 2011; Vergeynst et al., 2015), surface water (Blair et al., 2013; Carmona et al., 2014; Vergeynst et al., 2014), and sporadically in groundwater (Lapworth et al., 2012) and drinking water (Padhye et al., 2014) at concentrations ranging from the low ng L⁻¹ to µg L⁻¹. Pharmaceutical residues in the aquatic environment are reported to induce ecotoxic effects, hormonal disruption, and bacterial resistance (Fent et al., 2006; Kostich et al., 2014; Yan et al., 2014).

In developing countries and particularly in Africa, the information on the occurrence of pharmaceuticals in the environment is very scarce (Hughes et al., 2013; Aus der Beek et al., 2016) with, to the best of our knowledge, only few publications from Africa available in open literature (K'oreje et al., 2012; Agunbiade and Moodley, 2014; Manickum and John, 2014; Wood et al., 2015; Ngumba et al., 2016). Nevertheless, in most African countries, problems regarding both water quantity and quality are even more severe than in the developed regions. Kenya is a water scarce country facing serious water pollution challenges due to lack of proper (waste)water management and sanitation facilities. The national sewerage coverage is only 17% and varies within the main cities, e.g. Nairobi (28%), Mombasa (4%) and Kisumu (13%). Moreover, diseases are widespread amongst the population, and HIV/AIDS (29%), respiratory infections (14%) and malaria (6%) are the leading causes of deaths (Government of Kenya (2014)). Consequently, Kenya has a pharmaceutical consumption pattern with high use of antibiotics, analgesics and anti-inflammatory drugs, antimalarial compounds, and anti(retro)viral drugs at 13, 6, 5 and 4 ton year⁻¹ in Nairobi, respectively (K'oreje et al., 2012). However, the scarcity of data on the environmental occurrence of human and veterinary pharmaceuticals in use today impedes a proper estimation of exposure to pharmaceuticals in environmental and drinking water, and their potential risks to the ecosystem and human health (World Health Organization, 2012). This necessitates more research in these unexplored regions.

Therefore, the present study particularly focuses on concentrations and loads of multi-class pharmaceuticals in wastewater, river water and groundwater of two major Kenyan cities. For one of the first times, it also aims to provide some insight into the efficiency of typical wastewater stabilization ponds (WSPs) towards the removal of pharmaceutical residues.

2. Materials and methods

2.1. Description of the study area

The study was done in Nairobi and Kisumu city, Kenya (Fig. S1). Sampling sites were selected based on specific pollution sources. In Nairobi, samples were collected in September 2012 from the influent and effluent of the Dandora wastewater treatment plant (WWTP1) and its receiving River Ngong' (approx. 42 km long; 8 locations named RN1–8). In Kisumu, sampling was performed in July 2013 at the Nyalenda and Kisat WWTPs (WWTP2–3), and their receiving Rivers Auji (approx. 8 km long; 3 locations named RA1–3)

and Kisat (approx. 10 km long; 3 locations named RK1–3), respectively. Also three shallow wells located in an informal settlement (SW1–2) and rural area (SW3) in the Kisumu region have been sampled. WWTP1 and 2 are wastewater stabilization ponds (WSPs) consisting of a lagoon system comprising screening bars, anaerobic, facultative and maturation ponds, while WWTP3 operates through trickling filters. The treatment plants WWTP1, 2 and 3 have a hydraulic retention time of about 60–90 days, 6 days and 8 h, respectively. All the rivers are influenced by informal settlements, waste dump sites, industrial and WWTP effluent discharges. At each site, duplicate grab samples were collected in pre-cleaned 500 mL amber glass and 2 L plastic bottles for the analysis of pharmaceutical residues and physical-chemical parameters, respectively. The samples were transported to the laboratory in an ice-cooled box, stored at 4 °C in the dark, and extracted (SPE) within 3 days.

2.2. Analytical methods

The flow of the rivers at the sampling points was measured by the midsection method using a current meter and acoustic doppler velocimeter (Turnipseed and Sauer, 2010). The physical-chemical water quality was assessed based on pH, chemical (COD) and biochemical oxygen demand (BOD₅), turbidity, total suspended (TSS) and dissolved solids (TDS), conductivity, alkalinity and chloride concentration, using the Standard Methods for Examination of Water and Wastewater developed by the American Public Health Association (APHA, 2012). Pharmaceutical residues were analysed using solid-phase extraction and liquid chromatography coupled to magnetic sector high-resolution mass spectrometry, according to the method recently reported by Vergeynst et al. (2015). Briefly, water samples were loaded on Oasis HLB cartridges. After washing with water, the analytes were eluted with methanol and the extract was dried under nitrogen stream before reconstitution in 1 mL 10:90 methanol/water. Chromatographic separation was done using a Surveyor HPLC system. The binary mobile phase consisted of methanol (A) and water (B), both with 0.1% formic acid; and acetonitrile (C) and water (D) for ESI positive and negative ion mode, respectively. For detection, a double focusing magnetic sector MAT95XP HRMS was used, operating in multiple ion detection (MID) mode at a mass resolving power of 10 000 (10% valley definition). More details can be found in SI.

3. Results and discussion

3.1. Physical-chemical water quality parameters

In influent, COD and BOD₅ values range from 390 to 3200 mg L⁻¹ and 213–1750 mg L⁻¹, respectively (Table S1). Although these parameters were reduced by about 35–80% during wastewater treatment, the COD and BOD₅ effluent concentrations were up to 15–25 times higher than the Kenyan quality standards for effluent discharged into the environment. Similarly, the TSS and TDS (up to 140 mg L⁻¹ and 2175 mg L⁻¹, respectively) concentrations exceeded the standards in at least one WWTP and at several river sites. pH and chloride levels were in agreement to the standards, except for pH at river sites RK3, RA2 and RA3. The other measured parameters (turbidity, alkalinity and conductivity) are not included in the environmental legislation in Kenya.

The rivers show a high degree of organic pollution as evidenced by their COD (30–1278 mg L⁻¹) and BOD₅ (10–513 mg L⁻¹) concentrations, exceeding drastically the values (COD up to 18 mg L⁻¹; BOD₅ up to 4.3 mg L⁻¹) reported by e.g. Goher et al. (2014) and Yaping and Zongren (2012) in other developing countries like Egypt

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