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Pharmaceutical residues in water and sediment of Msunduzi River, KwaZulu-Natal, South Africa

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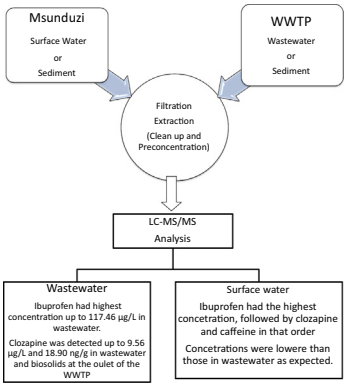
HIGHLIGHTS

- Data on pharmaceutical contamination of African water bodies is limited.
- We determined pharmaceutical residues in Msunduzi River in KwaZulu-Natal.
- Wastewater & bio-solids from a treatment plant in the catchment were also analysed.
- Residues were found in surface water, sediment, wastewater and bio-solids.
- The antipyretic ibuprofen exhibited the highest concentration in the samples.

GRAPHICAL ABSTRACT



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ABSTRACT

The little data about pharmaceutical residue contamination in African water bodies motivated our study on the occurrence of pharmaceutical residues in the water and sediment of Msunduzi River in the KwaZulu-Natal province of South Africa; and in the Darvill wastewater treatment plant found in Msunduzi catchment. Samples collected along the River and wastewater treatment plant were extracted and analysed for pharmaceutical residues selected based on statistics of drug usage in South Africa i.e. antipyretics, antibiotics, caffeine, an antiepileptic and an antipsychotic drug were determined using HPLC–MS/MS.

In all the matrices investigated, the antipyretic ibuprofen had the highest concentration of up to 117 µg L^{-1}, 84.60 µg L^{-1} and 659 ng g^{-1} in wastewater, surface water and sediment respectively. Antibiotics were detected in generally low concentrations of <10 µg L^{-1} in surface water samples and up to 34.50 µg L^{-1} in wastewater; moreover they were not completely removed during wastewater treatment. The percentage removal efficiency of the studied group was 6.55–98.00% for antipyretics, 73.33–98.90% for antibiotics, 48.80% for the anti-epileptic drug and 86.40% for Caffeine. Clozapine exhibited a negative removal.

In surface water, Henley dam exhibited a high concentration of the pharmaceutical residues and the highest concentration of metronidazole in sediment (up to 1253.50 ng g^{-1}) detected. Metronidazole was only detected in sediment and bio-solids.

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1. Introduction

Pharmaceuticals are known chemicals of environmental concern due to health risks associated with exposure of aquatic life to these compounds and possible risks to human health when they reach drinking water (Kümmerer, 2009b, 2010; Deblonde et al., 2011); therefore water sources should be monitored regularly. Several authors have reported the occurrence of pharmaceutical residues in waters in Europe, Asia and the USA (Chapman et al., 2002; Heberer, 2002; Löffler and Ternes, 2003; Kümmerer, 2009a; Fatta-Kassinos et al., 2011; Rehman et al., 2013; Manickum and John, 2013; Camacho-Muñoz et al., 2014; Luo et al., 2014) but there is limited information about their occurrence in African water bodies yet literature reports indicate that types of pharmaceuticals and/or their metabolites commonly detected are dependent on social, cultural, technological and agricultural factors (Dehghani et al., 2011; Agunbiade and Moodley, 2014) and therefore may be unique for different geographical areas and water bodies.

A study in Kenya reported occurrence of ibuprofen, paracetamol, sulfamethoxazole and zidovudine at a high concentration ($\sim$10–30 $\mu g\ L^{-1}$) in the Nairobi River basin (K'oreje et al., 2012) while studies on Umgeni River in KwaZulu-Natal, South Africa revealed presence of antibiotic, antipyretic, antiepileptic, antipsychotic drug residues and caffeine in surface water and sediment. While most of these pharmaceutical residues were <10 $\mu g\ L^{-1}$ in surface water, antipyretics were generally higher (Agunbiade and Moodley, 2014; Matongo et al., 2015). It is therefore important to study the occurrence and fate of pharmaceuticals in various water bodies because drug consumption patterns may differ by region. The consumption of pharmaceuticals in the Republic of South Africa for the period December 2012 to November 2013 as shown in Fig. 1 indicated that antipyretics were the most consumed followed by antibiotics. This consumption data was provided by ImpactRx Data Management (Pty) Ltd (http://www.impactrx.co.za/about-us) a South African company which reports pharmaceutical usage.

In this study we selected pharmaceutical residue targets basing on: annual consumption in South Africa, their persistent detection in water bodies world-wide as seen from the literature, their reported adverse effects and consideration to represent several therapeutic classes. Pharmaceutical residues were selected to represent the therapeutic classes of antipyretic, antibiotic, stimulant, antiepileptic and antipsychotic drugs because they were consumed in large amounts and also due to their reported ecotoxicological effects (Martin et al., 2012; Patrolecco et al., 2014).

Despite the various studies on the occurrence of pharmaceuticals in water bodies of Europe, Asia and USA, little data can be found in the literature concerning findings of pharmaceuticals in sediments, wastewater and surface water of African countries. In this study we investigated pharmaceutical residues in Msunduzi River which is found in South Africa. The quality of water of Msunduzi River (tributary length 115 km), an important tributary of Umgeni River in the KwaZulu-Natal Province of South Africa, is of concern. A recent investigation of its quality revealed microbial contamination with organisms associated with waste disposal such as *Salmonella* spp, *enterococci* and *E. coli* (Gemmell and Schmidt, 2013). This deterioration in quality attributed to increasing urbanization and large informal settlements within the Msunduzi catchment (size of 875 km^2), is associated with environmental problems such as contamination and pollution as a result of industrial waste, refuse dumping, and urban run-off.

The Msunduzi River runs through the Midlands of the KwaZulu-Natal (KZN) province in Eastern South Africa and includes a stretch through the provincial capital city of Pietermaritzburg. It joins the Umgeni River between Nagle and Inanda dams and flows out into the Indian Ocean at Durban (http://www.arocha.org/int-en/work/sites/past/g5/945-DSY.html). It is a source of water for domestic agricultural and industrial use in the Msunduzi Municipality. In a recent study on the Umgeni, pharmaceutical residues were detected at a higher concentration at the sampling point where Msunduzi River joins Umgeni (Matongo et al., 2015).This observation motivated investigation of the Msunduzi River to determine whether it may be contributing to the pharmaceutical loading of the Umgeni.

The discharge of insufficiently treated municipal wastewater into rivers has been identified as a major pathway responsible for surface water contamination with pharmaceuticals and personal care products (PPCPs); some pharmaceutical compounds are not completely removed by conventional treatment processes and eventually end up in drinking water (Okuda et al., 2009). Therefore we investigated Darvill a wastewater treatment plant

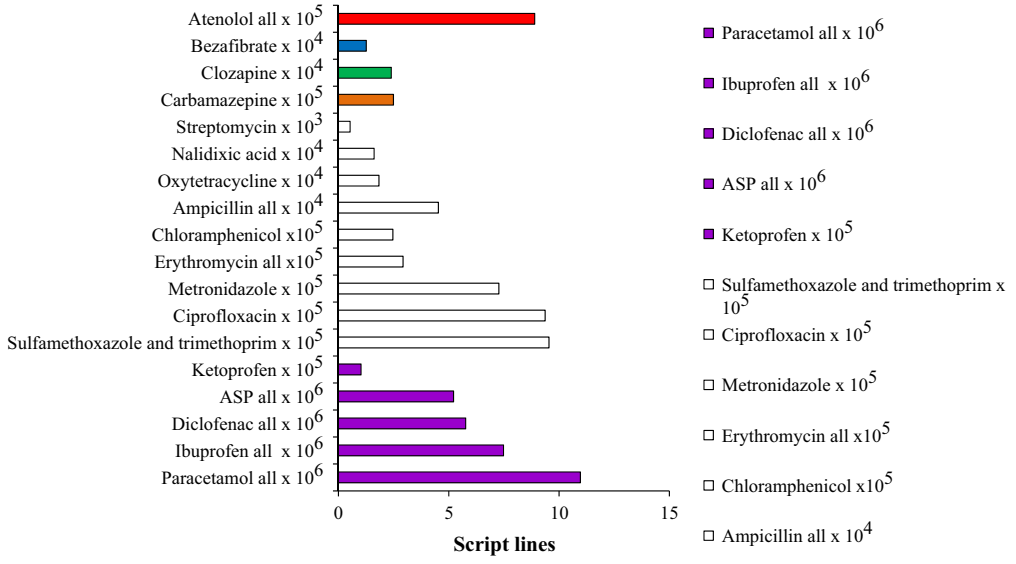

Fig. 1. Script lines in RSA. Paracetamol all includes all paracetamol and paracetamol combinations with or without psycholeptics, ibuprofen all includes all ibuprofen and ibuprofen combinations with or without psycholeptics. Similarly diclofenac includes all diclofenac and diclofenac combinations with or without psycholeptics.