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‘An investigation into the occurrence and removal of pharmaceuticals in Colombian wastewater’

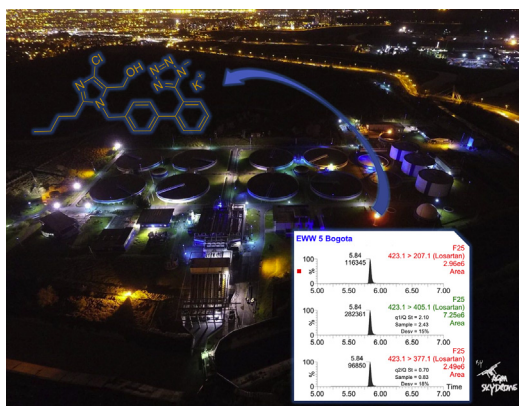
A.M. Botero-Coy [a], D. Martínez-Pachón [b], C. Boix [a], R.J. Rincón [b], N. Castillo [b], L.P. Arias-Marín [c], L. Manrique-Losada [d], R. Torres-Palma [e], A. Moncayo-Lasso [b,*], F. Hernández [a,*]

[a] Research Institute for Pesticides and Water (IUPA), University Jaume I, Castellón, Spain
[b] Grupo de Investigación en Ciencias Biológicas y Químicas, Facultad de Ciencias, Universidad Antonio Nariño, Bogotá D.C., Colombia
[c] Grupo Bioprocesos Microbianos (Biomicro), Escuela de Microbiología, Universidad de Antioquia UdeA, Calle 70 No. 52-21, Medellín, Colombia
[d] Grupo de Investigación en Materiales, Ambiente y Desarrollo (MADE), Facultad de Ciencias Básicas, Universidad de la Amazonia, Florencia, Colombia
[e] Grupo de Investigación en Remediación Ambiental y Biocatálisis (GIRAB), Instituto de Química, Facultad de Ciencias Exactas y Naturales, Universidad de Antioquia UdeA, Calle 70 No. 52-21, Medellín, Colombia

HIGHLIGHTS

- Pharmaceuticals were present at ppb levels in urban and hospital wastewaters from Colombia.
- Effluent wastewater from conventional WWTPs contained concentrations similar to raw influent wastewater.
- Both IWW and EWW are directly discharged in the aquatic ecosystem in some locations.
- Removal efficiency of pharmaceuticals in WWTPs is incomplete, therefore additional tertiary treatments are needed.
- The use of LC-MS/MS QqQ allowed reliable quantification supported by quality control analysis.

GRAPHICAL ABSTRACT



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ABSTRACT

In this work, the presence of 20 pharmaceuticals in wastewater from Colombia is investigated. Several widely consumed compounds have been detected in wastewater samples from different origins and geographical areas in Colombia. The studied pharmaceuticals included antibiotics, analgesics and anti-inflammatories, cholesterol lowering statin drugs, lipid regulators, and anti-depressants. The investigated samples were urban wastewater collected during one whole week before (influent) and after treatment (effluent) in the wastewater treatment plants (WWTPs) of Bogotá and Medellin. Raw wastewater from the Hospital of Tumaco and from the city of Florencia were also collected. Analyses performed by liquid chromatography-tandem mass spectrometry (LC-MS/MS) revealed that most of the target analytes were present in all the wastewater samples. The highest concentrations (up to 50 μg/L) corresponded to acetaminophen, but several antibiotics, such as azithromycin, ciprofloxacin and norfloxacin, and antihypertensive drugs, such as losartan and valsartan, were commonly present in influent wastewater (IWW) at levels above 1 μg/L. Moreover, the treatment applied in WWTPs seemed to not efficiently remove the compounds under study, because most pharmaceuticals were also present in effluent wastewater (EWW) at concentrations close to those of the IWW. Special emphasis was made in this work on the quality of data reported, performing a detailed study of quality control (QC) samples.

* Corresponding authors.
E-mail addresses: amoncayo@uan.edu.co (A. Moncayo-Lasso), felix.hernandez@uji.es (F. Hernández).

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The analytical approach used –direct injection of 5-fold diluted samples without any additional treatment – is simpler and faster than the commonly applied solid phase extraction (SPE). The use of 12 isotope-labelled internal standards ensured the satisfactory correction of matrix effects for the corresponding analytes. For the remaining 8 compounds, no drastic matrix effects were observed, and only four compounds (cloxacillin, doxycycline, losartan, tetracycline) presented QC recoveries near or slightly below 60%, revealing ionization suppression, particularly in the IWW. Data on the occurrence of pharmaceuticals reported in this paper are the basis for current studies that aim to develop efficient systems for the degradation/removal of these compounds from the aquatic environment.

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1. Introduction

Currently, there is special concern about the presence and potential hazardous effects of emerging contaminants (EC) in the aquatic environment. ECs are widely used compounds that are not regulated yet (or only a few of them); however, they are commonly found in wastewater and surface water at concentrations that may be higher than classical persistent and/or priority substances; therefore, they can be harmful to aquatic ecosystems and become a hazard for human public health (Daughton, 2004; Gracia-Lor et al., 2012a; Hernández et al., 2015a; la Farré et al., 2008). Within the wide group of ECs, pharmaceuticals are among the most frequently detected compounds in the aquatic environment (Hughes et al., 2013). Antibiotics are of particular concern due to the potential risks associated with the development of microorganisms resistant to antibiotics (Makowska et al., 2016; Manaia et al., 2016). The evaluation of the hazards of emerging contaminants, such as pharmaceuticals and personal care and disinfection by products, is a current priority in regulatory water quality monitoring (Loos et al., 2009; Brack et al., 2012; Brack et al., 2017).

It was approximately 40 years ago when pharmaceuticals were first considered environmental contaminants in the aquatic environment (Hignite and Azarnoff, 1977; Richardson and Bowron, 1985). Several pioneer works highlighted the interest in studying the occurrence and impact of pharmaceuticals in the aquatic environment (Daughton and Ternes, 1999; Daughton, 2004; Kolpin et al., 2002). From then, there has been an impressive increase in the number of publications reporting the presence of pharmaceuticals in the water environment. The development of sophisticated analytical techniques played a key role in their emergence (Hernández et al., 2015b; Richardson and Kimura, 2016). Data reported by many environmental laboratories around the world reveal that pharmaceuticals are omnipresent in the aqueous environment (e.g., wastewater influent and effluent, manure, industrial effluent, surface water, groundwater, and drinking water (aus der Beek et al., 2016; Monteiro and Boxall, 2010)). Liquid chromatography coupled to tandem mass spectrometry (LC-MS/MS) has played a prominent role in this process. LC-MS/MS is the most widely applied technique for the determination of emerging contaminants in water, due to its excellent sensitivity, selectivity and robustness, together with reliability in the identification of the compound detected. For the successful application of LC-MS/MS, at least two MS/MS transitions need to be monitored and their ion ratios evaluated (Hernández et al., 2018; García-Galán et al., 2016; Gracia-Lor et al., 2014; Bayen et al., 2014; van Nuijs et al., 2010; Pozo et al., 2006a; Pozo et al., 2006b).

As expected from the wide human consumption, the main source of pollution for pharmaceuticals is urban wastewater, and it is rather common to find high pharmaceuticals concentrations (up to hundreds of ppb) in influent wastewater (IWW). Unfortunately, conventional wastewater treatment plants (WWTP) do not efficiently remove these compounds, and they can remain in effluent wastewater (EWW) at concentrations similar, or even higher, than in IWW (Gracia-Lor et al., 2012a; Gros et al., 2010; Jelic et al., 2011; Lacey et al., 2008). In addition to the inefficient removal in WWTPs, some pharmaceuticals are also persistent and can bioaccumulate in living organisms (Daughton and Ternes, 1999; Yang et al., 2014). As a consequence, these compounds can easily reach surface water (Dai et al., 2015; Kasprzyk-Hordern et al., 2008; Fatta-Kassinos et al., 2011; Liu et al., 2015; Matamoros et al., 2012) and even drinking water (Boyd et al., 2003; Carmona et al., 2014; Kumar and Xagoraraki, 2010; Sodré et al., 2010; Vulliet et al., 2011).

Although much data are available around the world, revealing the presence of pharmaceuticals in different aquatic scenarios, little information is available from Latin America. This is a particular worry, because in some areas, the discharge of raw sewage into rivers, lakes and reservoirs is rather common (De Paula et al., 2007; Gracia-Lor et al., 2012b; Montagner and Jardim, 2011; Thomas et al., 2014). Concern over the presence of these contaminants is well founded, because these water resources not only flow through tropical areas rich in biodiversity, but surface waters are also often used as a source for human consumption (Aristizabal-Ciro et al., 2017).

Recently, we performed a wide-scope qualitative screening of organic contaminants, including pharmaceutical, veterinary drugs, X-ray agents, personal care products, sweeteners, illicit drugs, and a notable number of metabolites in wastewater and receiving surface waters from the area surrounding Bogotá (Hernández et al., 2015a). The use of high resolution MS, specifically liquid chromatography coupled to quadrupole-time-of-flight (LC-QTOF MS) allowed us to identify a notable number of pharmaceuticals in both EWW and in surface water, emphasizing acetaminophen, carbamazepine, clarithromycin, diclofenac, lincomycin, losartan and valsartan, among others, together with several metabolites. As a consequence of that work, future monitoring, based on quantitative analyses, was recommended to focus on those compounds found in the screening.

In this work, we have performed a quantitative analysis, based on LC-MS/MS with triple quadrupole, for selected pharmaceuticals in Colombian wastewater. Raw wastewater samples from the Hospital of Tumaco (Nariño), as well as from the city of Florencia (Caquetá), which are currently discharged directly in two rich biodiversity regions in Colombia (the Pacific and Amazonian regions), were analysed. In addition, influent and effluent wastewater samples collected during one week in the cities of Bogotá and Medellin (department of Antioquia) were also analysed. The aim of this work was 1) to estimate the removal efficiency of WWTPs from analysis of IWW and EWW; 2) to have better knowledge on the compounds present in effluent wastewater that potentially reaches the surface waters receiving discharges from WWTPs; 3) to allow the design of complementary treatments, such as the advanced oxidation processes, that we are investigating at present for the efficient removal of the pharmaceuticals most commonly found in wastewater.

2. Experimental

2.1. Characteristics of the wastewater treatment plants and studied areas

The analysed samples were collected in two of the main cities of Colombia (Bogota D.C. and Medellin, department of Antioquia), in the young city located in southeast Colombia (Florencia – Caquetá) and in one coastal pacific city (Tumaco). In the case of Bogotá and the department of Antioquia, a total of 28 samples were taken from the

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