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Pollution by psychoactive pharmaceuticals in the Rivers of Madrid metropolitan area (Spain)

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ABSTRACT

There are a number of reports in the literature which describe the occurrence of so-called emerging pollutants, such as pharmaceuticals, in surface water. Few of these studies have reported values from large cities in relatively arid areas, such as in Spain. The persistence of some pharmaceuticals to usual wastewater treatments allows their discharge into surface waters. It is increasingly evident that mental health problems are of special concern for public health since psychiatric drugs are used in large amounts. Compared to other countries, Spain has a high pharmaceutical consumption rate, and Madrid metropolitan area is one of the most densely populated areas in Europe. The aim of the present study was to determine the presence of different psychoactive pharmaceuticals and metabolites in the main rivers of Madrid metropolitan area: Jarama, Manzanares, Guadarrama, Henares and Tajo. Sampling was done downstream of ten sewage treatment plants (STP) discharging into these rivers. Control points upstream of STPs discharge points were also sampled. Pharmaceutical compounds and metabolites for analysis were selected according to human consumption and prescription rates in Madrid, and the availability of valid techniques for detection. We observed residues of the antidepressants fluoxetine (80% of the sampling sites), citalopram (60%) and venlafaxine (100%), the anxiolytics nordiazepam (90%), oxazepam (80%) and 7-aminoflunitrazepam (10%) and the anticonvulsant carabamazepine (70%). Measured concentrations equalled or exceeded those reported for other geographical areas, although there is a pronounced lack of information for the anxiolytics and venlafaxine. This is of special concern given that Wyeth–Ayerst's venlafaxine, Effexor, was the 10th greatest selling pharmaceutical worldwide in 2006. We conclude that the origin of pharmaceutical pollution in the rivers of Madrid is mainly the discharge of sewage treatment plants in Madrid's metropolitan area and a comprehensive monitoring program should be implemented.

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1. Introduction

In the past years a reduction in the amount of "conventional" pollutants in surface water (nitrates, heavy metals, sulphur and nitrogen oxides, etc.) has occurred worldwide, but we are witnessing the occurrence of the so-called "emergent" pollutants, which include pharmaceuticals and their metabolites (Fent et al., 2006). Low concentrations of human pharmaceuticals have been detected in several countries, either in sewage treatment plants (STPs), surface waters, sea water, groundwater, sediments or drinking water. These studies have been performed in central and eastern Europe, Scandinavia, Mediterranean countries, South and North America and Asia (reviewed in Glassmeyer et al., 2008). In Spain, several studies on the efficacy of STPs to remove pharmaceuticals have been performed with new, more sensitive analytical methods (Agüera et al., 2003; Carballa et al., 2004; Carballa et al., 2008; Gomez et al., 2007; Gros et al., 2006; Lamas et al., 2004; Martínez Bueno et al., 2007; Morales-Muñoz et al., 2005; Radjenovic et al., 2007; Santos et al., 2005). However, there are few studies on the occurrence of pharmaceuticals in the environment and the ones that exist are exclusively focused in the north eastern corner of the Iberian Peninsula (Gros et al., 2007, 2009; Huerta-Fontela et al., 2008; Kuster et al., 2008; Lamas et al., 2004).

The published reports show that the pharmaceuticals present in the Rivers of Madrid belong to five main therapeutical groups: analgesics–

Abbreviations: C, control; CID, collision induced dissociation; CM, Community of Madrid; CNS, central nervous system; DSSTP, downstream sewage treatment plants; ESI+, electrospray in the positive ionization mode; IS, internal standards; PNEC, predicted no-effect concentrations; SSNRIs, selective serotonin and norepinephrine reuptake inhibitor; STP, sewage treatment plants.

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antipyretics, anti-inflammatories, antibiotics, antilipemic agents and β-blockers. Some of them, like the anticonvulsant carbamazepine or the analgesic diclofenac occur usually in the 100s of ng/L, but can sometimes occur in the μg/L range, with values varying from one country to another in the effluents of STPs (reviewed by Zhang et al., 2008).

Some of the studies recently published state that low concentrations of many of the pharmaceutical drugs found do not constitute an imminent risk for public health (Khetan and Collins, 2007). However, some ecotoxicological studies show evidence of chronic toxicity for aquatic organisms due to a continuous exposure to low doses (Ternes et al., 1999; Stackelberg et al., 2004; Fent et al., 2006; Khetan and Collins, 2007). Special attention should be paid to studies about the effect of mixtures because of possible interactions among different pharmaceuticals (Cleuvers, 2003; Cleuvers, 2004; Fent et al., 2006).

Spain is currently the World's 7th largest consumer of pharmaceuticals, despite being ranked 29th in terms of population (IMS Health 2006; http://www.imshealth.com). Spain spent more than 14×10^9€ on pharmaceuticals in 2008, which equals around 300€ *per capita*. The most consumed therapeutical groups in Spain are those related to cardiovascular and central nervous system pathologies (CNS) (Farmaindustria, 2004). Likewise it is increasingly evident that mental health problems are a growing concern for public health. In Europe, according to an investigation published in 2004, the prevalence of psychiatric diseases in the general population was 11.1% (Alonso et al., 2004). In the last few years, the number of people with these disorders has increased. Problems relating to mental health are a priority of the European Commission according to the Public Health Program (2003–2008). Antidepressants, antipsychotics, and anticonvulsants are some of the top selling pharmaceutical drugs through retail pharmacies (October 2008) ranking third, fourth and fifth respectively (IMS Health 2008; http://www.imshealth.com).

In spite of the high consumption of pharmaceuticals and the high population density of Madrid, no study has determined the levels of psychoactive pharmaceutical drugs in the surface waters of Madrid. The aim of the present study was to determine the presence of different psychoactive pharmaceuticals and metabolites in the main rivers of the Madrid metropolitan area.

2. Material and methods

2.1. Sampling site

The Community of Madrid (CM) is located in central Spain (Fig. 1), with a total area of 8021.80 km^2 and an estimated population of 6,081,689 inhabitants (13.45% of the population of Spain, National

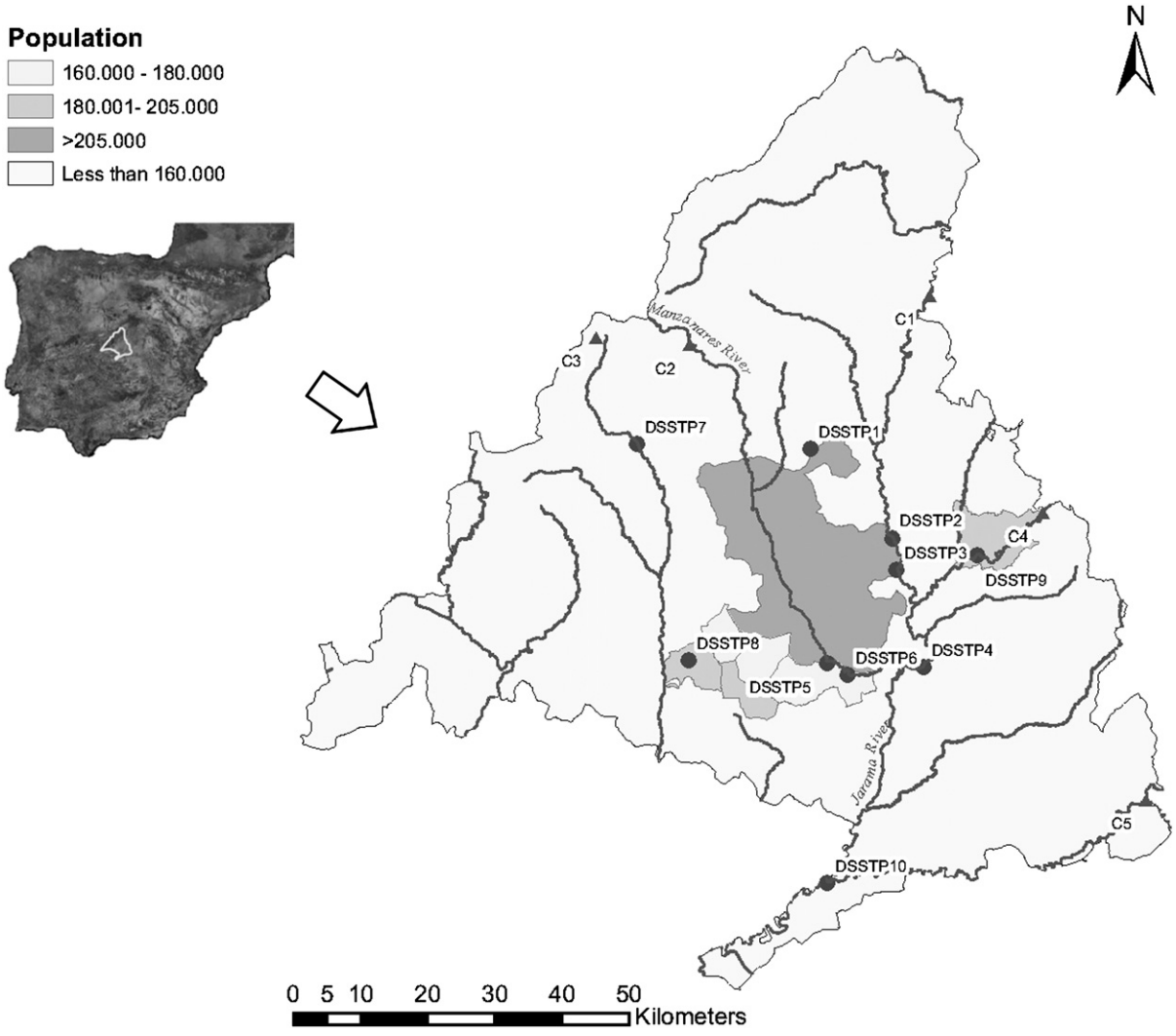

Fig. 1. Map of the study area and sample site locations (DSSTP and C) in the Community of Madrid.

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