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A comparative assessment of intensive and extensive wastewater treatment technologies for removing emerging contaminants in small communities

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ABSTRACT

Ecosystem pollution due to the lack of or inefficient wastewater treatment coverage in small communities is still a matter of great concern, even in developed countries. This study assesses the seasonal performance of 4 different full-scale wastewater technologies that have been used in small communities (<2000 population equivalent) for more than 10 years in terms of emerging contaminant (EC), chemical oxygen demand (COD), total suspended solids (TSS) and NH_4–N removal efficiency. The studied technologies, which were selected due to their widespread use, included two intensive treatment systems (an extended aeration system (AS) and a rotating biological contactor (RBC)) and two extensive treatment systems (a constructed wetland (CW) and a waste stabilization pond (WSP)), all located in northeastern Spain. The studied compounds belonged to the groups of pharmaceuticals, sunscreen compounds, fragrances, antiseptics, flame retardants, surfactants, pesticides and plasticizers. The 25 ECs occurred in wastewater at concentrations ranging from undetectable to 80 $\mu g\ L^{-1}$. The average removal efficiency was 42% for the CW, 62% for the AS, 63% for the RBC and 82% for the WSP. All the technologies except the WSP system showed seasonal variability in the removal of ECs. The ecotoxicological assessment study revealed that, whilst all the technologies were capable of decreasing the aquatic risk, only the WSP yielded no risk in both seasons.

1. Introduction

Although almost all wastewater generated in developed countries is treated, for a large part of the population living in small communities (<2000 population equivalent, PE), sewage is still generally poorly treated or even untreated (Molinos-Senante et al., 2014). In fact, according to recent studies (Aragón et al., 2013), sanitation and wastewater treatment coverage in small communities in countries such as Spain is less than 50%, and it is estimated that a huge number of small wastewater treatment plants (WWTPs) should be built in the near future to provide a long-term solution. Moreover, since most of these communities are located in remote areas, the sewage is usually discharged into rivers or streams with a high biodiversity value. In such cases, special attention is required to improve wastewater treatment and identify the best available treatment technology in order to achieve a good chemical and ecological status of the surface waters, as set out in the Water Framework Directive (Directive 2000/60/EC). Although considerable information can be found on these wastewater technologies' performance with regard to nutrient removal (Molinos-Senante et al., 2012), there is a lack of information on their ability to remove specific pollutants such as emerging contaminants (ECs), as well as on the adverse ecotoxicological effects that the presence of such compounds can have on surface water bodies.

Numerous chemical compounds we use in our daily lives enter the environment, where they are considered ECs. They include a wide variety of compounds, such as pharmaceuticals, personal care products, plasticizers, flame retardants, surfactants and certain pesticides, amongst others. Since conventional WWTPs are not designed to treat these types of contaminants, many of these compounds occur at different concentrations in natural water bodies (Jiang et al., 2013; Ternes et al., 2004), where they can exert ecotoxicological effects even at relatively low concentrations (Henry and Black, 2008; Muñoz et al., 2009). Some of the known environmental effects of the presence of ECs in surface waters are the reduction of macroinvertebrate diversity in rivers (Muñoz et al.,

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2009) and behavioural changes in mosquito fish (Henry and Black, 2008). Different ecotoxicological approaches have been used to assess the effect of the presence of these compounds in surface water bodies, but the simplest strategy remains the evaluation of the hazard quotient (HQ) between the chemical composition of the water samples and the predicted no-effect concentration (PNEC) for different water organisms (EPA, 1998).

Since the main source of ECs in surface water bodies is the discharge from WWTPs (Luo et al., 2014), it is very important to identify the most efficient treatment technologies. Basically, two main approaches for wastewater treatment can be identified: intensive and extensive. Although the most commonly used intensive technology in small WWTPs is the extended aeration system (AS), which is a modification of the conventional activated sludge system but with a higher hydraulic retention period (Aragón et al., 2013; Tsagarakis et al., 2000), other intensive technologies such as rotating biological contactors (RBCs) or tricking filters (TFs) are available. For example, in a review study, Luo et al. (2014) showed that the AS technology was able to remove some ECs by up to 90%. In a laboratory-scale study, Vasiliadou et al. (2014) observed that an RBC was capable of removing recalcitrant compounds such as carbamazepine (88%). In contrast to these intensive technologies, in recent years the use of extensive systems, such as constructed wetlands (CWs) or waste stabilization ponds (WSPs), has attracted considerable interest due to their better landscape integration as well as their low maintenance costs (Polprasert and Kittipongvises, 2011) and the greater economic, environmental and social sustainability (Molinos-Senante et al., 2014). CWs can be of different configurations, but the vertical and horizontal flow CWs are the most common. The main difference between them is that while the former works under prevalent aerobic conditions the latter works under anaerobic conditions. In Spain, the most commonly used is the horizontal configuration. As has been reported elsewhere, CWs and WSPs are as efficient at removing ECs as conventional activated sludge technologies (Matamoros et al., 2013). Li et al. (2013) reported that, except for carbamazepine, the WSP technology's removal efficiencies for the other detected pharmaceuticals and personal care products were relatively high, with an overall removal efficiency ranging from 88 to 100%. Similarly high removal efficiencies have been reported for CWs under aerobic conditions (Matamoros et al., 2007). Although different studies have been conducted on the effectiveness of intensive and extensive technologies for removing ECs, most were performed on a pilot scale, whilst others failed to take seasonality into account. Therefore, a survey study addressing these issues is still needed.

This study aimed to evaluate the seasonal occurrence and removal efficiency for 25 ECs in four different intensive and extensive small wastewater treatment technologies widely used in north-eastern Spain (i.e. a CW, an AS, a WSP, and a RBC). The selected WWTPs were full-scale systems which have been serving small communities for more than 10 years under the same operational conditions. The ECs belonged to the groups of pharmaceuticals, sunscreen compounds, fragrances, antiseptics, fire retardants, surfactants, pesticides and plasticizers. Finally, an aquatic risk assessment study was performed based on the HQs.

2. Material and methods

2.1. Description of the small WWTPs

The treatment technologies were selected on the basis of being the most used technologies in the Catalan territory, being in operation for more than 10 years without a recorded accident. They were representative of each of the studied technologies, which was proved by recorded data on conventional wastewater parameters (BOD_5, TSS and NH_4–N) with other WWTPs using the same technology (information provided by the Water Catalan Agency). The studied treatment technologies were as follows: a CW located in Sant Martí de Sesgueioles (550 PE), with a design capacity of 150 $m^3\ d^{-1}$; an AS located in Sant Martí de Tous (968 PE), with a design capacity of 240 $m^3\ d^{-1}$; a WSP located in Sant Guim de Freixenet (956 PE), with a design capacity of 200 $m^3\ d^{-1}$; and an RBC located in Viladecavalls (2000 PE), with a design capacity of 650 $m^3\ d^{-1}$. Fig. 1 shows a schematic design of each of the studied treatment technologies. The WSP consisted of three ponds, two of which were connected in parallel with a surface area of 4500 m^2 each. These two ponds were fed alternatively every 15 days. The water effluent from the ponds was treated in a final polishing pond with a surface area of 1500 m^2. The maximum water depth in all the ponds was 2 m, with a HRT of around 20–30 days. The AS consisted of a primary clarifier and an aerated reactor integrated with a secondary clarifier with a total surface area of 80 $m^2$and a HRT of 2 days. The CW consisted of two horizontal subsurface flow cells, spanning 600 m^2 and 1000 m^2 respectively. Both cells were filled with gravel, had a water depth of 0.5 m and were planted with *Phragmites australis*. The two wetland cells were fed intermittently with a HRT of 4–6 days. Finally, the RBC consisted of a primary clarifier and two rotating cylinders that were 4 m in diameter and approximately 10 m long. The treated wastewater was decanted by means of a secondary clarifier with a surface area of 50 m^2 and with a HRT of 1–2 days. All the WWTPs also made use of a primary treatment: either sedimentation tanks or clarification tanks, with a hydraulic residence time (HRT) higher than 24 h.

2.2. Sampling strategy

Two sampling campaigns were carried out, the first in July 2013 (warm season) and the second in February 2014 (cold season). In each campaign, grab water samples were collected from the influent and effluent of all the treatment systems every day for a week. No rainfall events were recorded at any time during the sampling period. No differences in the wastewater inflow were observed between campaigns for each of the studied WWTPs. Due to the difficulty of setting up an automatic composite sampler, water samples were always collected after the primary treatment, which was used as a homogenization tank. All the studied WWTPs included a primary treatment (either a sedimentation tank or a clarification reactor), which was set at a HRT higher than 24 h. This helped reduce the huge influent variability normally observed in raw wastewater composition. All the water samples were collected in 1000 mL amber glass bottles, which were transported under refrigeration to the laboratory, where they were stored at 4 °C until analysis. The sample holding time was less than 12 h.

2.3. Analytical procedures

Conventional wastewater quality parameters, including ammonium nitrogen (NH_4–N), total suspended solids (TSS) and chemical oxygen demand (COD), were determined in all the water samples. The COD was measured with Hach Lange COD cell tests (LCK 314 and LCK 614) on a spectrophotometer (Hach Lange Pocket Colorimeter II at 450 nm). Onsite measurements of water temperature and dissolved oxygen (DO) were taken using a Checktemp-1 Hanna thermometer and a Eutech Ecoscan DO6 oxygen meter, respectively. Chemicals and reagents used for the analytical procedures are provided in the supplementary material (SM) section.

All the water samples were filtered and processed as previously reported (Matamoros and Bayona, 2006). A 100 mL sample was spiked with 50 ng of a surrogate standard (atrazine D5, mecoprop D3, tonalide D3, and dihydrocarbamazepine). The spiked sample