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Balancing environmental quality standards and infrastructure upgrade costs for the reduction of microcontaminant loads in rivers

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ABSTRACT

Investments for upgrading wastewater treatment plants (WWTPs) with tertiary treatment to reduce microcontaminant loads in surface waters at a catchment scale can be daunting. These investments are highly sensitive to the selection of environmental quality standards (EQSs) for the target micro-contaminants. Our hypothesis is that there is a balance between EQS selection and investment that needs to be considered in decision-making. We used a customized microcontaminant fate and transport model coupled to an optimization algorithm to validate this hypothesis in the Llobregat river basin and for the pharmaceutical compound diclofenac. The algorithm optimizes the number of WWTPs in this catchment requiring an upgrade to minimize the total amount of diclofenac that exceeds the EQS in every river section and the total cost. We simulated and optimized 40 scenarios representing a combination of 4 potential EQSs (10, 30, 50 and 100 ng L^{-1}), 5 levels of uncertainty bounds in the predictions of river concentrations and 2 hydrological scenarios (average flows, flows annually exceeding 30% of the days; and environmental flows, flows annually exceeding 99% of the days). The results showthat there is a nonlinear relationship between the EQS and the required investment. The investment increases by 100% from an EQS of 100 ng L^{-1} to 10 ng L^{-1}, significantly increasing (by 60%) from 30 to 10 ng L^{-1}. Thus, establishing an EQS of 30 ng L^{-1} would balance environmental protection and costs. The selection of the hydrological conditions also plays a key role in the upgrade analysis because the costs for environmental flows are 50% higher than for average flows. Finally, we highlight that the investment in research would allow the reduction of uncertainties, hence allowing more qualified decisions to be made and a reduction in the WWTP upgrade costs (up to 4 €·$household^{-1}\cdot year^{-1}$).

1. Introduction

The presence of microcontaminants in surface waters raises environmental and human health concerns and has become a key environmental problem (Acuña et al., 2015a). To protect human health and aquatic life, environmental agencies, countries and territories establish environmental quality standards (EQSs) for these contaminants. The establishment of EQSs for micro-contaminants results from a scientific process (based on ecotoxicological studies) and a political process (i.e. in the European Union (EU), amendments to the EQS Directive 2008/105/EC are negotiated in the European Council and the European Parliament; European Commission, 2012). Even though there are some guidelines available (European Commission, 2011b), the approaches on how to set EQS values differ among countries and territories (European Commission, 2012; Ecotox Centre, 2017). Taking diclofenac as an example, several EQSs have been proposed in Europe, ranging from 10 ng L^{-1} (European Medicines Agency, 2006) to 100 ng L^{-1} (European Commission, 2012). The selection of EQS should be fully consistent with the precautionary principle, but the economic implications surrounding the establishment of an EQS are not fully understood. Investments for upgrading wastewater treatment plants (WWTPs; from secondary biological treatment –most commonly - to tertiary treatment) to reduce microcontaminant loads in surface waters can be daunting. For the capital and operating costs, Hillenbrand et al. (2014) estimated that the upgrade of all the German WWTPs serving more than 5000 population equivalent (PE) would cost approximately 1.3 billion € annually

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(3013 WWTPs requiring an upgrade for a total of 82 M inhabitants in Germany). The Swiss Federal Office for the Environment (BAFU, 2012) estimated that the upgrade of the required 123 WWTPs would cost 1 billion € in total (0.12 billion € annually, for a total of 8.5 M inhabitants in Switzerland). Provisional estimates by the UK government showed that the upgrade of 1360 WWTPs in England and Wales would cost between 32 and 37 billion € in total (Owen and Jobling, 2012). Our hypothesis is that there is a balance between EQS selection and investment that needs to be considered in decision-making. For diclofenac, the number of European river stretches exceeding a potential EQS increases exponentially as the EQS decreases from 100 ng L^{-1} to 10 ng L^{-1} (Johnson et al., 2013; Kehrein et al., 2015). We expect a similar relationship between the potential EQS and the corresponding investment to avoid EQS exceedance.

National/regional water agencies have the responsibility of allocating resources for upgrading WWTP infrastructure. Cost-effective allocation implies optimizing resources at a catchment scale and can take advantage of models. Such optimization approaches have already been demonstrated in the research field, as seen in Bishop and Grenney (1976), Udias et al. (2012), and Saberi and Niksokhan (2017), but are mostly applied to the reduction of conventional contaminants (organic matter, ammonia, nitrate, etc.).

Such an optimization assessment has never been conducted for microcontaminants. A few studies evaluated (and some optimized) the implementation of strategies to decrease microcontaminant concentrations in rivers below the EQSs; however, none of them conducted a proper economic assessment. For instance, Ort et al. (2009) optimized the number of WWTPs (but not the cost) to be upgraded in Switzerland to avoid any exceedance of the diclofenac EQS. Coppens et al. (2015) prioritized the number of WWTPs (not optimizing or assessing the cost) to be upgraded in the Netherlands based on the impact of pharmaceutical emissions on drinking water and ecology. Gimeno et al. (2017) evaluated several interventions implemented at every WWTP in a Spanish catchment (not optimizing or assessing the cost) to avoid exceedance of the diclofenac EQS. Finally, Kehrein et al. (2015) evaluated the influence of EQS selection on the number of stretches showing non-compliance in the Ruhr watershed (Germany). Therefore, the trade-off between EQS selection and costs remains unknown.

Hence, this paper aims to evaluate the relationship between the potential EQS for diclofenac and the cost of the WWTP upgrades required to avoid EQS exceedance. This is done through a model-based optimization of the WWTP upgrade costs for different EQSs in a case study in the Llobregat River basin (Catalonia, Spain). Ozonation is selected as the upgrading technology for the removal of microcontaminants. This study also evaluates the influence of the selection of river discharges and of the model uncertainty on the optimization of the WWTP upgrade costs.

2. Materials and methods

2.1. Study area and target compound

The study area is the Llobregat River basin, which is the second longest river in Catalonia (Spain). The main axis of the river extends 165 km from the Pyrenees to the Mediterranean Sea, draining an area of 4948 km^2, and has two main tributaries, the Cardener and Anoia Rivers. The hydrology of the Llobregat River is characterized by a highly variable flow that is strongly influenced by seasonal rainfall. The mean annual bulk precipitation is 3330 hm^3, and it has an annual average bulk discharge of 693 hm^3. The basin includes 56 WWTPs (54 conventional activated sludge, 1 aerated lagoon and 1 membrane bioreactor, with PE ranging from 100 to 280,000), which collect and treat wastewater from 1.1 M inhabitants (Statistical Institute of Catalonia, 2016) and discharge to the Llobregat (Fig. 1). Our target compound is diclofenac, a common non-steroidal anti-inflammatory drug. Diclofenac has been shown to bioaccumulate in fish and invertebrates at environmentally relevant concentrations (Huerta et al., 2015) and to potentially exert harmful effects on non-target aquatic organisms at higher concentrations (Acuña et al., 2015b). Diclofenac has been included on the EU “watch list” of priority substances under the Water Framework Directive (Directive 2013/39/EU, European Commission, 2013; Decision (EU) 2015/495; European Commission, 2015). There is no definitive EQS for diclofenac (European Commission, 2011a), so it is an appropriate compound for this study.

2.2. Microcontaminant fate and transport model

We used the microcontaminant fate and transport (MFT) model developed in Gimeno et al. (2017) to describe the fate and removal of diclofenac along the entire Llobregat River basin. The tool integrates 3 submodels: 1) a substance-human consumption and excretion model, which estimates the diclofenac loads that reach the influents of WWTPs; 2) a WWTP model; and 3) a river model. Each submodel has a key parameter: 1) F is a lumped factor that includes the fraction of the diclofenac parent compound that is excreted to toilets, discharged directly via sinks and washed off of skin or clothes; 2) k_{WWTP} is the reaction rate constant that incorporates processes by which diclofenac is degraded; and 3) k_{river} is the reaction rate constant that represents natural diclofenac degradation in rivers. The model was calibrated as in Gimeno et al. (2017) using measurements of diclofenac in WWTP influents and effluents and in the river during September 2010. The calibrated model parameters values are shown in Table 1. The output of the MFT model is a calibrated probability distribution function (PDF) of predicted diclofenac concentrations in every river stretch in September 2010. We refer to Gimeno et al. (2017) for further details on the MFT development and calibration.

The model of Gimeno et al. (2017) was expanded to include ozonation after secondary wastewater treatments. Ozonation is able to almost completely remove the diclofenac present in secondary effluents (95–99%) at a low ozone dose (Hollender et al., 2009). The estimated cost for ozone appears to be lower than other technologies, such as UV and activated carbon (Wahlberg et al., 2016; Mulder et al., 2015). However, harmful by-products are generated during ozonation, so we also considered a filtration step (sand filter) afterwards (Hollender et al., 2009). The percentage of diclofenac removal through ozonation and sand filtration is described by the coefficient α in equation (1). Hence, the diclofenac load from secondary effluent (after conventional activated sludge treatment, L_{eff}) simulated by the MFT model would be additionally removed by (100-α)/100. The load of diclofenac after ozonation and sand filtration is depicted as L_{tert}. We assumed that this technology could only be installed at WWTPs larger than 5000 PE (18 of 56 WWTPs in the catchment). Installing ozonation in WWTPs smaller than 5000 PE is not feasible because ozonation requires qualified permanent staff for their operation (Rossi et al., 2013). Moreover, the sum of PE corresponding to the WWTPs smaller than 5000 PE only represents 6% of the total PE in the Llobregat basin. We have set α to 99 because diclofenac removal is 99% for the ozone dose assumed in this study (0.7 g $O3 \cdot g\ DOC^{-1}$; Zimmermann et al., 2011, Hollender et al., 2009).

$$L_{tert} = L_{eff} * (100 - \alpha)/100 \quad (1)$$

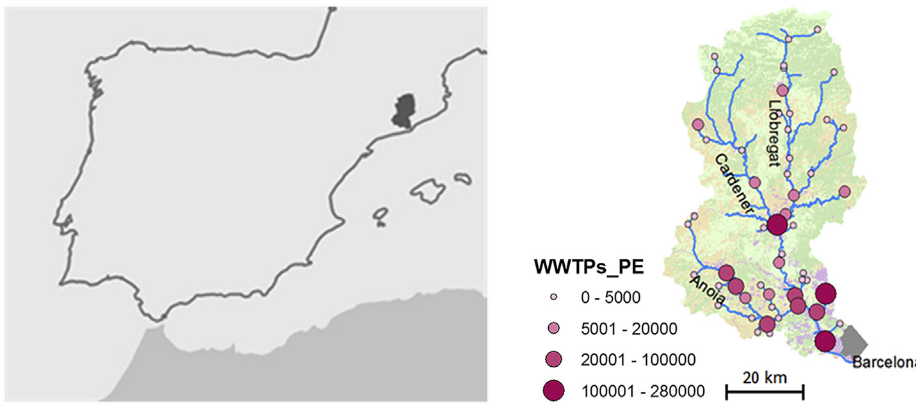

Fig. 1. Left: Location of the Llobregat basin in Spain. Right: The Llobregat River catchment, main tributaries (Cardener and Anoia) and location of WWTPs (magenta circles). WWTPs are ranked based on the population equivalent served (extracted from Gimeno et al., 2017).

Table 1
Calibrated F, k_{WWTP} and k_{river} values for diclofenac in September 2010 as in Gimeno et al. (2017).

	5^{th} percentile	Median	95^{th} percentile
F (dimensionless)	0.11	0.15	0.23
k_{WWTP} ($l \cdot gss^{-1} \cdot d^{-1}$)	0.12	0.25	0.70
k_{river} (s^{-1})	1.4E-07	3.0E-06	1.5E-05

2.3. Ozonation costs

We collected the yearly costs (capital and operational) of 11 ozonation systems followed by sand filtration from literature (Mulder et al., 2015; Hunziker, 2008; Abegglen et al., 2009; Margot et al., 2013; Biebersdorf, 2014). While the capital costs include investment, realization and project costs, the operational costs account for personnel, maintenance and variable costs. The variable costs include the electrical consumption for ozone generation and sand filtration and the cost of pure oxygen for the ozone production (Table 2). We assumed that ozone is generated from pure oxygen instead of from ambient air. This is justified because almost fivefold higher ozone concentrations can be generated from pure oxygen and about half energy is consumed when ozone is generated from oxygen instead of air (Gottschalk et al., 2010). The assumption is that the ozone dosage is 0.7 $g\ O3 \cdot g\ DOC^{-1}$ and the retention time in the ozonation tank is 25 min, which is the lowest ozone dose considered in Mulder et al., (2015) to calculate the ozonation costs. These ozonation operating conditions allow reaching a removal of 99% or higher of diclofenac (Zimmermann et al., 2011; Hollender et al., 2009).

The approach proposed in Mulder et al. (2015) was applied to obtain a full estimate of costs for each of these 11 systems. Thus, we calculated the yearly investment costs assuming a lifetime of 30 years for civil works and 15 years for machinery and electrical equipment and a yearly interest rate of 4% (as well applicable to Spain; Spanish Central Bank, 2010). For the ozonation systems which did not provide yearly capital costs, we applied an increase of 65% (the ratio between investment and project and realization costs from Mulder et al., 2015) to the investment costs to account for realization and project costs. The yearly maintenance costs are calculated as 3.5% of the total investment costs as in Mulder et al. (2015). These calculations are included in the Supporting Information – excel file "ozonation capital costs". Personnel and variable costs were adjusted to the reality in Spain (Table 2), hence accounting for Spanish salaries and the price of electricity. We obtained the salary of a qualified operator in WWTPs from the Spanish Ministry of Employment and Social Security (BOE, 2017). We obtained the price of electricity from Eurostat (2017). The price of electricity in Spain in 2017 for non-domestic consumers decreases as the yearly use increases (0.135 $€ \cdot kWh^{-1}$ for a use between 20 and 500 $MWh \cdot year^{-1}$; 0.101 $€ \cdot kWh^{-1}$ for a use between 500 and 2000 $MWh \cdot year^{-1}$ and 0.084 for a use between 2000 and 20,000 $MWh \cdot year^{-1}$; Eurostat, 2017). These values include an expected increase of 5.3% in the price of electricity by 2050 (European Commission, 2016). The cost of pure oxygen also varies depending on the treatment capacity. We used a cost of 0.15 $€ \cdot kg^{-1}$ for ozonation systems that treat less than 750 $m^3\ h^{-1}$ of wastewater (Prieto-Rodríguez et al., 2013) and 0.08 $€ \cdot kg^{-1}$ for ozonation systems that treat more than 750 $m^3\ h^{-1}$ (Ried et al., 2009). The calculations to obtain the variable costs are included in the Supporting Information – excel file "ozonation variable costs". We highlight in green in Table 2 those values that were extracted from literature and used directly in our study. We highlight in blue those values that were estimated in this study. The rest of the specifications in Table 2 were extracted from Mulder et al. (2015).

We obtained the cost function using the costs in Table 2 and fitted them to a power function so that we can estimate the cost for any ozonation treatment size (equation (2)), where PE accounts for the population equivalent. We included the goodness of fit of the cost values to the potential function in Supporting Information - Figure SI-1. For the WWTPs smaller than 11,000 PE (minimum WWTP size with cost of ozonation in this study), we assumed that the ozonation costs increase following the same power function as in equation (2). This was justified by the use of one single power function that fits real ozonation costs in WWTPs ranging from 5000 to 1,000,000 PE as in Hillenbrand et al. (2014) and Roccaro et al. (2013).

$$Cost\left(€ \cdot m^{-3}\right) = 6.824 \times (\mathrm{PE})^{-0.344} \tag{2}$$

2.4. Optimization of the number of WWTP to be upgraded

We used the Non-dominated Sorting Genetic Algorithm II (NSGA-II; Deb et al., 2002) implemented in Matlab® (The Mathworks, Inc.) to find the optimal set of WWTPs that should be upgraded to minimize the cost and EQS exceedance accumulated in all river stretches of the catchment. Hence, we defined two objective functions: (I) minimization of the total yearly cost of the upgrades (eq (3)) and (II) minimization of the total load of diclofenac exceeding EQS (eq (4)).

Table 2

Breakdown of the costs per m^3 treated effluent of ozonation followed by sand filtration. The capital costs of ozonation for 14,000, 70,000 and 210,000 PE were extracted directly from Mulder et al. (2015). The investment costs of ozonation for 11,000, 45,000, 57,000, 120,000 and 500,000 PE were provided by Hunziker (2008). Abegglen et al. (2009), Margot et al. (2013) and Biebersdorf (2014) provided the investment costs of ozonation for 35.000, 30.000 and 74.000 PE respectively. The yearly investment costs from Hunziker (2008), Abegglen et al. (2009), Margot et al. (2013) and Biebersdorf (2014) were increased by 65% to account for the realization and project costs. The yearly maintenance costs were calculated as 3.5% of the total investment. The salary of 1 qualified operator was obtained from the BOE (2017). The prices of electricity were obtained from Eurostat (2017) and the price of pure oxygen from Prieto-Rodríguez et al. (2013) and Ried et al. (2009). We highlight in green those values that were extracted from literature and used directly in our study. We highlight in blue those values that were estimated in this study. The capital and variable cost breakdowns are included in the Supporting Information – excel "ozonation capital costs" and "ozonation variable costs".

Capacity (PE)	11,000	14,000	30,000	35,000	45,000	57,000	70,000	74,000	120,000	210,000	500,000
Design capacity post treatment ($m^3 \cdot h^{-1}$)	130	180	360	430	550	710	900	930	1400	2700	6000
Treated volume ($m^3 \cdot year^{-1}$)	759,200	1,024,920	2,102,400	2,511,200	3,212,000	4,146,400	5,124,600	5,431,200	8,176,000	15,373,800	35,040,000
Capital costs (€·year^{-1})	170,000	140,000	270,000	150,000	330,000	470,000	590,000	380,000	550,000	1,570,000	1,100,000
Investment costs											
-Technical life time: civil works (30 years), machinery and electrical equipment (15 years)											
-Interest: 4%											
Realization and project costs: 65% of investment											
-Engineering (12%), insurances, permits and other building costs (15%), project management and construction supervision (8%), temporary installations (5%), training personnel (2%), communication (2%), VAT (21%)											
Maintenance (€·year^{-1}): 3,5% of investment	29,000	22,000	43,000	27,000	58,000	79,000	100,000	56,000	93,000	220,000	184,000
- Civil works (0.5%), machinery and electrical equipment (3%)											
Personnel costs (€·year^{-1})	8300	8300	8300	8300	16,700	16,700	16,700	16,700	25,000	25,000	25,000
Small WWTP - 1/3 qualified operator salary											
Medium WWTP - 2/3 qualified operator salary											
Large WWTP - 1 qualified operator salary											
Variable costs (€·year^{-1}), including 21% VAT)	32,000	47,000	89,000	106,000	136,000	149,000	137,000	151,000	227,000	440,000	863,000
- Electricity: 0,135 €·kWh^{-1} for 20–500 MWh·year^{-1}; 0,101 €·kWh^{-1} for 500- 2000 MWh·year^{-1}; 0,084 €·kWh^{-1} for 2000–20,000 MWh·year^{-1}											
- Pure oxygen: 0.15 €·kg^{-1} for design flow < 750 $m^3 \cdot h^{-1}$ and 0.08 €·kg^{-1} for flow > 750 $m^3 \cdot h^{-1}$											
Total yearly cost (€·year^{-1})	239,300	217,300	410,300	291,300	540,700	714,700	843,700	603,700	895,000	2,255,000	2,172,000
Cost (€·m^{-3})	0.32	0.21	0.20	0.20	0.17	0.17	0.16	0.11	0.11	0.15	0.06

$$Min\sum_{i=1}^{N} Cost \quad (3)$$

$$Min\sum_{i=1}^{M}((Conc \times Q_{stretch}) - (EQS \times Q_{stretch}) \quad (4)$$

where N is the number of WWTPs to be upgraded with ozonation and sand filtration, M is the number of stretches with EQS exceedance, *Conc* represents the predicted concentration of diclofenac in the river stretch, and $Q_{stretch}$ is the flow simulated in the stretch.

Since there are only 18 WWTPs within the Llobregat River basin with more than 5000 PE, we selected 18 discrete variables "α" to be optimized either with a value "99" (reflecting that ozonation was installed after that WWTP and 99% of diclofenac was removed before being discharged to rivers) or with a value "0" (reflecting that ozonation was not installed in that WWTP so diclofenac is not further removed). For the rest of the WWTPs in the Llobregat, diclofenac was not further removed (only the removal given by the conventional activated sludge process - average value of 38%; Gimeno et al., 2017). Regarding the NSGA-II parameters, we selected the population size and the number of generations following a "trial and error" approach and ensuring that we evaluate the extreme objective function values (minimum cost and maximum exceedance, and maximum cost and minimum exceedance). Consequently, the population size ranged between 200 and 300, and the number of generations ranged between 100 and 150, depending on the scenario evaluated (section 2.5). The result of the optimization is the "Pareto front" (see example in Supporting Information - Figure SI-2). The "Pareto front" shows the cost of the upgrades and EQS exceedance of every solution (which includes a particular set of WWTPs) at each generation. The optimal solutions are plotted in the last generation. We selected the optimal solution that minimizes the EQS exceedance the most to compare costs and the number of WWTPs requiring an upgrade between scenarios. We ran the optimization algorithm for each scenario as described hereafter.

2.5. Simulation of scenarios of an EQS under different hydrology and uncertainty levels

We combined 4 different EQSs, 2 hydrological conditions and 5 levels of uncertainty. Hence, in total, we optimized the set of WWTPs to be upgraded for 40 scenarios. We ran the optimizer NSGA-II for each scenario, and we selected the optimal solution that minimizes the EQS exceedance the most for each level of uncertainty and hydrological scenarios.

We evaluated the EQS of 10, 30, 50 and 100 ng L^{-1} in surface waters proposed for diclofenac. We believe that we covered very different levels of environmental protection considering a wide range of EQSs. In 2012, the European Commission (EC) suggested an EQS of 100 ng L^{-1} for diclofenac (European Commission, 2012). However, noting that this value could be under protective, the EC suggested that this value had to be reviewed later on, taking into account the lowest observed effect concentrations (LOECs) and producing other reliable studies. In 2017, the Swiss Centre for Applied Ecotoxicology suggested an EQS of 50 ng L^{-1} (Ecotox centre, 2017) based on the NOEC in fish determined by Birzle (2015). In addition, Acuña et al. (2015a) suggested a value of 30 ng L^{-1}, which corresponded to the 5th percentile of the LOEC for aquatic biota. Furthermore, the European Medicines Agency (2006) fixed a threshold safety value of 10 ng L^{-1} in the environmental risk assessment (ERA) procedures for pharmaceuticals. New ecotoxicity data has to be determined in the future concerning chronic effects and mixtures of chemicals, and the EQS for mixtures may be preferable to deriving EQSs for the individual constituent substances (Kienzler et al., 2016). Overall, there is no agreement in the definition of an EQS for diclofenac.

As for the hydrological conditions, we considered *average flows* (those measured in September 2010) and *environmental flows* (minimum flows in the Llobregat River). September 2010 is the period that was used for data collection and model calibration in Gimeno et al. (2017). The river flows of September 2010 correspond to the average hydrological conditions in the Llobregat. Considering the series of daily flows measured over the last 10 years (flow monitoring stations in Supporting Information - Figure SI-3), the river flows of September 2010 correspond to $Q_{30\%}$ (flow exceeded 30% of the days in 10 years). The *environmental flows* were determined by the Catalan Water Agency (Official Journal of the Government of Catalonia, 2006) under the principles of progressive implementation and compatibilization of environmental needs and existing uses, with special attention given to safeguarding supply guarantees. This environmental flow regime is defined for all bodies in the district, especially for the flows in the Llobregat river basin which are mainly controlled by a system of upstream reservoirs. The Catalan Water Agency is currently using the environmental flows to assess the compliance of wastewater discharges with environmental standards. Hence, the Catalan Water Agency suggested using the environmental flows as the minimum flows in this study. The Catalan Water Agency is also taking measures to ensure these *environmental flows* in their rivers, even during severe droughts. Considering the series of daily flows measured over the last 10 years (flow monitoring stations in Supporting Information - Figure SI-3), the *environmental flows* correspond to $Q_{99\%}$ (flow exceeded 99% of the days in 10 years).

We evaluated the scenarios of calibrated and reduced model parameters (F, k_{WWTP}, and k_{river}) uncertainty (Gimeno et al., 2017). For the scenario of calibrated model parameter uncertainty, we optimized the cost of the WWTP upgrades using the calibrated diclofenac concentrations in the Llobregat in September 2010. For the scenario of reduced model parameter uncertainty, we used the diclofenac concentrations that were simulated with reduced parameter uncertainty with respect to the calibrated uncertainty (simulating the MFTM with reduced parameter uncertainty (i.e., 60% reduction with respect to the calibrated uncertainty) leads to reduced uncertainty in diclofenac concentrations (Gimeno et al., 2017)). For each scenario, we evaluated the highest, median and lowest probable concentrations as in Johnson et al. (2013). The median concentrations are identical in both scenarios of uncertainty. Thus, we evaluated 5 levels of uncertainty in diclofenac concentrations. We simulated the highest probable concentrations using the 95th percentile of F and the 5th percentile of k_{WWTP} and k_{river}, respectively, for each scenario; the median probable concentrations using the 50^{th} percentile of the 3 model parameters and the lowest probable concentrations using the 5th of F and the 95th percentile of k_{WWTP} and k_{river}. We assumed the same calibrated and reduced PDFs of model parameters for both hydrological conditions. However, we expect a higher removal of diclofenac during *environmental flows* because we considered lower velocities in the stretches for these low flows (Kunkel and Radke, 2012).

3. Results

3.1. Influence of different EQSs on the cost of the upgrades

As expected, the total annual cost of the upgrades reduces as the EQS increases, and this is consistent for both hydrological scenarios

(Fig. 2 and Fig. 3). For the scenario *average flows* (Fig. 2), we obtained a non-linear relationship between EQS and the cost of the upgrades (negative power relationship, see goodness of fit in Supporting Information - Figure SI-5). The cost to avoid EQS exceedance varied from 10.1 M€·year^{-1} (14 WWTPs requiring upgrade for EQS of 10 ng l^{-1}) to 4.8 M€·year^{-1} (5 WWTPs requiring upgrade for EQS of 100 ng l^{-1}), a difference of almost 6 M€·year^{-1} (median values). The highest decrease in costs was found between 10 ng L^{-1} and 30 ng L^{-1} (from 10.1 M€·year^{-1} to 6.2 M€·year^{-1}, respectively). For the scenario *environmental flows* (Fig. 3) the cost varied linearly from 11.1 M€·year^{-1} to 8.8 M€·year^{-1} (median values for different EQS). The differences in cost among EQS 30 ng L^{-1} and 50 ng L^{-1} were lower than 1 M€·year^{-1} for both hydrological scenarios (approximately 0.2 M€·year^{-1} for *average flows* and approximately 1 M€·year^{-1} for *environmental flows*). The sets of WWTPs that are upgraded under each EQS optimization are included in the Supporting Information - Figures SI-4 and SI-5.

3.2. Influence of hydrological conditions on the cost of the upgrades

Higher upgrade costs would be required to avoid EQS exceedance under *environmental flows* compared to *average flows* (median values). While the median cost of the upgrades to comply with an EQS of 100, 50 and 30 ng L^{-1} is lower than 6.5 M€·year^{-1} for *average flows*, the median cost is always higher than 8.5 M€·year^{-1} for *environmental flows*. Indeed, the cost increased by 84% for an EQS of 100 ng L^{-1}, by 67% for 50 ng L^{-1} and by 77% for 30 ng L^{-1} for *environmental flows* compared to *average flows*. The number of upgraded WWTPs under *environmental flows*, being more than twice the number under *average flows*, explains those increases. Nearly the same optimal set of WWTPs to be upgraded is obtained under both hydrological conditions if the EQS was 10 ng L^{-1}.

3.3. Influence of uncertainty on the cost of the upgrades

The uncertainty in diclofenac concentrations in the river resulted from simulating the model using the 5th, 50th and 95th

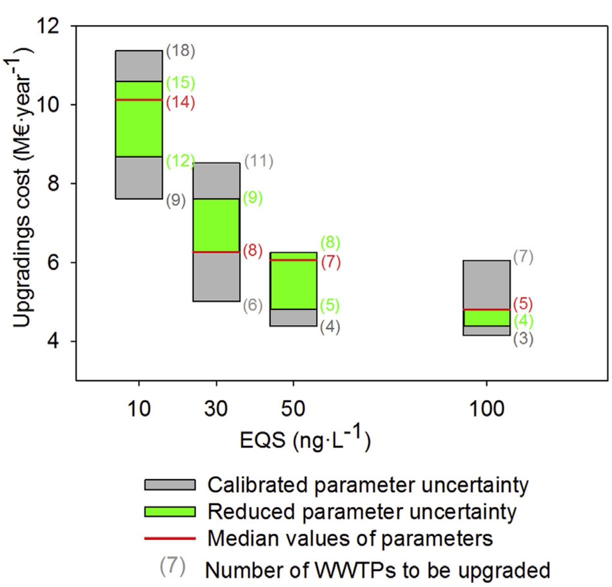

Fig. 2. Optimal cost of the required WWTP upgrades with ozonation to reduce diclofenac concentrations in rivers below an EQS of 10, 30, 50 and 100 ng L^{-1} during *average flows* and for the calibrated and reduced model parameter uncertainty. The optimal number of WWTPs to be upgraded is shown for the highest, median and lowest probable concentrations of diclofenac.

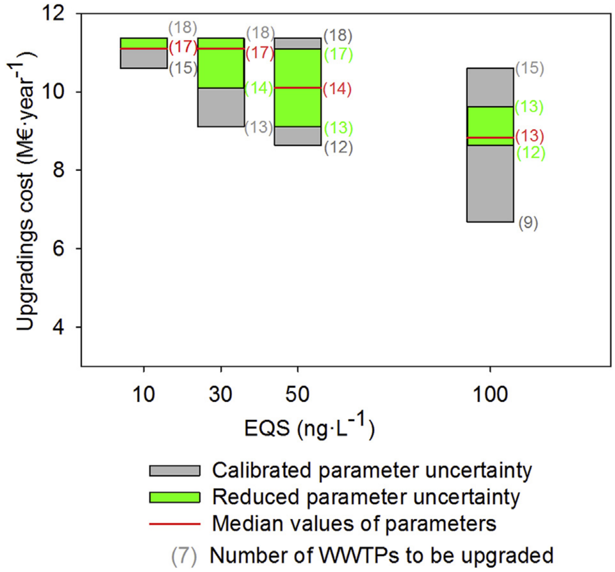

Fig. 3. Optimal cost of the required WWTP upgrades with ozonation to reduce diclofenac concentrations in rivers below an EQS of 10, 30, 50 and 100 ng L^{-1} during *environmental flows* and for the calibrated and reduced model parameter uncertainty. The optimal number of WWTPs to be upgraded is shown for the highest, median and lowest probable concentrations of diclofenac.

percentiles of the parameters (see section 2.5). Such uncertainty entails variability in the cost of the upgrades for every EQS and scenario. For the calibrated uncertainty, the variability in the cost ranges from 2% (0.3 M€·year^{-1} for an EQS of 10 ng L^{-1} and *environmental flows*) to 36% (2.3 M€·year^{-1} for an EQS of 30 ng L^{-1} and *average flows*). The variability in the cost is larger as the EQS decreases under the *average flows* scenario (from 1.9 M€·year^{-1} for an EQS of 100 ng L^{-1} to 3.7 M€·year^{-1} for 10 ng L^{-1}), but the opposite is observed under the *environmental flows* scenario (from 0.8 M€·year^{-1} for 10 ng L^{-1} to 3.9 M€·year^{-1} for 100 ng L^{-1}). In the scenario of *average flows*, this is justified by an increase in the number of river stretches exceeding the lower EQS, and therefore, additional WWTPs are likely to be upgraded. Conversely, for *environmental flows*, concentrations of diclofenac are exceeding the lower EQS in almost every river stretch. Thus, nearly every WWTP should be upgraded, hence explaining the lower variability in cost for the lower EQS.

The variability in the cost of the upgrades decreases for every EQS and scenario when the model uncertainty is reduced. The variability in the cost (interquartile range) decreases from 23% (for an EQS of 50 ng L^{-1} and *average flows*) to 78% (for an EQS of 100 ng L^{-1} and *average flows*). Most likely, decision-makers would use the highest probable concentrations of diclofenac (highest value observed for each box plot) to make a conservative decision. We observe that the costs of the highest probable concentrations decrease when the model uncertainty is reduced. This means, that reducing uncertainty leads to a solution with decreased costs. As an example, for an EQS of 100 ng L^{-1} and *average flows*, we obtain a reduction in the cost of the upgrades of 1.3 M€·year^{-1} if the model uncertainty is reduced. Surprisingly, considering the calibrated uncertainty, the lower probable cost of the upgrades to avoid exceedance of a more stringent EQS (e.g., 30 ng L^{-1} and *average flows*) could be much lower than the median cost required for a less stringent EQS (e.g., 50 ng L^{-1} and *average flows*). We obtained more accurate solutions and costs considering reduced uncertainty (e.g., the lowest probable cost to avoid 30 ng L^{-1} exceedance and *average flows* is indeed higher than any probable cost to avoid 50 ng L^{-1}

exceedance). This is explained by the more accurate concentrations of diclofenac simulated using the reduced uncertainty compared to the calibrated uncertainty. These accurate concentrations cause the upgrade of further WWTPs to avoid 30 ng L^{-1} exceedance compared to 50 ng L^{-1} exceedance for average flows.

Finally, we observed that there is always a set of 3 WWTPs (Rubí, Terrassa and Sant Feliu) that is included in every optimal solution regardless of the EQS, uncertainty and hydrological scenario (Supporting Information - Figure SI-6). Thus, an investment of 4.1 M€·year^{-1} is required in any scenario for upgrading these 3 WWTPs.

4. Discussion

4.1. Innovation of the study: relationship between the EQS and the costs of the WWTP upgrades

The results confirm our hypothesis that the cost of the upgrades is highly sensitive to the potential EQS (from more than 10 M€·year^{-1} for an EQS of 10 ng L^{-1} to 5 M€·year^{-1} for an EQS of 100 ng L^{-1} and *average flows*), significantly increasing for the lowest EQS. The relationship between the EQS and costs becomes non-linear (negative power relationship, Supporting Information - Figure SI-7) for *average flows*, and hence, the cost of the upgrades to avoid 10 ng L^{-1} increases rapidly compared to 30 ng L^{-1} (from 6 M€·year^{-1} to more than 10 M€·year^{-1}). This is explained by the discrete nature of the optimization variables (WWTPs that are optimized can either be upgraded and removing diclofenac by an extra 99% or not). In this study, a small decrease in the EQS (from 30 to 10 ng L^{-1}) involves the need for upgrading a significantly higher number of WWTPs (from 8 to 14). The relationship between the EQS and the cost of the upgrades is useful for policy-makers when establishing cost-effective EQSs for microcontaminants and for decision-makers (e.g., Catalan Water Agency) when proposing interventions to comply with those EQSs. In the derivation of an EQS, given the non-linearities, European policy-makers should consider the daunting cost of the upgrades required to avoid exceedance of the more stringent EQS (i.e., 10 ng L^{-1}). Ort et al. (2009), Hillenbrand et al. (2014) and Kehrein et al. (2015) evaluated the required interventions at the WWTPs by minimizing diclofenac concentration exceedance for a single EQS of 100 ng L^{-1}. However, these studies did not evaluate the compliance with other proposed EQSs nor optimize the number of the WWTP upgrades to minimize costs and EQS exceedance. Thus, this is the first study that searches for the trade-off between the cost of the upgrades and compliance with the EQS for microcontaminants.

4.2. Comparison to existing national strategies for the reduction of microcontaminants in rivers

This section illustrates the advantages of our methodology with respect to other referenced criteria or methods. First, the criteria used to define the optimal number of WWTPs to be upgraded with advanced treatment for the removal of microcontaminants is catchment-dependent, while the use of our model helps river basin authorities (RBA) to find the optimal set of WWTP to be upgraded within any catchment. This is illustrated by implementing the Swiss strategy for the upgrade of WWTPs (BAFU, 2012) on the Llobregat River basin. The Swiss strategy proposed the upgrade of every WWTP serving up to more than 80.000 residents (microcontaminant load reduction), WWTPs serving up to more than 24.000 residents discharging into lakes (drinking water protection) and WWTPs serving up to more than 8.000 residents that contribute to more than 10% of the dry-weather stream flow (low river dilution capacity). This strategy was based on the modeling results from Ort el al. (2009) considering an EQS of 100 ng L^{-1}. Following this strategy (considering *environmental flows* as the dry-weather stream flow), 8.3 M€·year^{-1} should be invested to upgrade 10 WWTPs in the Llobregat basin. However, by upgrading this set of WWTPs, the median concentrations of diclofenac still exceed the EQS of 100 ng L^{-1} in 21 river stretches. The solution given by the Swiss strategy seems to be risky compared to any solution that we optimized for the *environmental flows*. For nearly the same cost, we avoid 100 ng L^{-1} exceedance during *environmental flows* by upgrading 13 WWTPs (median values). Hence, the upgrade of additional WWTPs in the Llobregat and higher costs are required to comply with the more stringent EQS and considering the uncertainty of diclofenac concentrations. On the other hand, lower cost (4.8 M€·year^{-1}) and fewer upgraded WWTPs (only 5) are required to avoid exceedance of 100 ng L^{-1} considering the median concentrations and *average flows* and compared to the solution given by the Swiss strategy. This means that the set of 10 WWTPs resulting from the Swiss strategy is not the optimal solution for the Llobregat river basin considering the minimization of both the cost of the upgrades and the EQS exceedance. This also suggests that uniform criteria for the selection of WWTPs to be upgraded across Europe would not be suitable for all countries given differences in hydrological conditions, treatment levels, etc.

Our methodology also helps RBAs prioritize which set of WWTPs should be first upgraded to avoid any EQS exceedance. There is always a set of WWTPs (Rubí, Terrassa and Sant Feliu) that is included in every optimal solution regardless of the EQS, uncertainty and hydrological scenario (Supporting Information - Figure SI-6). The effluents of these 3 WWTPs discharge to river stretches with very low river dilution capacity (wastewater contributing to more than 40% of the river flow). This explains the very high diclofenac concentrations simulated just downstream these plants (higher than 400 ng L^{-1}) that far exceeded any EQS. Therefore, this is the first study on prioritizing investments at WWTPs for the removal of microcontaminants considering both EQS compliance and costs.

4.3. Framing the optimal solutions into current operational costs and European experiences

The current operational cost of the WWTPs discharging into the Llobregat is 16.8 M€·year^{-1} (Catalan Water Agency, 2017). Considering that the operational cost of the upgrades (personnel, maintenance and variable costs) represents 40% of the total required cost (Table 2), the current operational cost would increase from 10% (considering the upgrade of 3 WWTPs with a total cost of 4.1 M€·year^{-1}) to 27% (upgrade of 18 WWTPs with a cost of 11.4 M€·year^{-1}). The Catalan Water Agency finances the required investment at the WWTPs through a water tax (*cànon de l'aigua*) that is included in each household water bill (Catalan Water Agency, 2017). For an average water use between 108 and 188 m^3·household^{-1}·year^{-1} (second tax block with lower water use) in Catalonia in 2016, the water tax was 102 €·household^{-1}·year^{-1}, and the total water bill (including water supply, wastewater, the water tax and VAT) was 355 €·household^{-1}·year^{-1} (Catalan Water Agency, 2016). Assuming that every household in the Llobregat contributes to the payment of the WWTP upgrades, the cost estimated in this study would represent an increase in the household water bill from 10 to 28 €·household^{-1}·year^{-1}. In percentages, this means an increase from 10% to 28% in the water tax and an increase from 3% to 8% in the total water bill. Assuming that the average household's income in Spain in 2017 is 40,000 €·household^{-1}·year^{-1} (OECD, 2017), the cost of the upgrades ranges from 0.3‰ to 0.7‰ of the household's income. The estimated average willingness to pay per household for upgrading the WWTPs in Switzerland (86 €·household^{-1}·year^{-1}; Logar et al., 2014) involves an increase by 20% in the total water bill in Switzerland (430 €·household^{-1}·year^{-1}; Logar et al., 2014) and just 1.2‰ of the average household's income in 2017 (66,000 €·household^{-1}·year^{-1}; OECD, 2017). Therefore, the cost of the upgrades represents a lower percentage of the household's water bills and income compared to the estimated WTP in Switzerland.

The cost of the upgrades in the Llobregat can be compared with the cost of the upgrades estimated in Switzerland (0.12 billion € annually for the upgrade of 123 WWTPs, as estimated in Logar et al., 2014) and Germany (1.3 billion € annually for the upgrade of 3013 WWTPs, as in Hillenbrand et al., 2014). Assuming that the cost is covered by every household's water bill in Switzerland and Germany, the required upgrades would mean an increase of 37 and 40 €·household^{-1}·year^{-1}, respectively. These values are larger than the [10–28] €·household^{-1}·year^{-1} estimated for the Llobregat River basin. The costs in Switzerland and Germany might be reduced if a model-based optimization (using real costs) would be applied. Finally, the decrease in parameter uncertainty could lead to savings down to 1.3 M€·year^{-1} in the selection of the optimal set of WWTPs to be upgraded and a reduction in the water tax up to 4 €·household^{-1}·year^{-1}. If a cost-benefit analysis is carried out to support research projects aimed to reduce uncertainties in MFT model parameters, the reduction in the cost of the upgrades can be incorporated in the analysis as a monetary benefit.

4.4. Use of hydrological conditions for decision-making on the removal of microcontaminants in rivers

In this study, we provide evidence that the cost of optimal interventions varies from 10% to 84% when using *average flows* or *environmental flows*, depending on the EQS. So far, there is no agreement on which river flows should be used for decision-making on the selection of measures to reduce micro-contaminant levels at a basin scale. Hillenbrand et al. (2014) evaluated WWTP interventions to decrease concentrations of 12 chemicals, including 4 pharmaceuticals, in the Neckar River basin (Germany) using the annually mean flow from 2008 to 2010. Likewise, Kehrein et al. (2015) considered the mean flow of the period 2012–2014 to evaluate measures to reduce diclofenac below EQS in the Ruhr River basin (Germany). Coppens et al. (2015) prioritized the investments at the WWTPs to reduce concentrations of carbamazepine and ibuprofen in Dutch rivers using the average flow of the driest and wettest 3-month period out of ten years (1996–2006). Ort et al. (2009) optimized the number of Swiss WWTPs to be upgraded for the removal of diclofenac for the $Q_{95\%}$ river flows (flow exceeded 95% of the time, annually averaged over a ten-year period). Kumar et al. (2014) evaluated the compliance of the estrogen E1 and E2 concentrations with standards in the Yodo River (Japan) for $Q_{50\%}$ and $Q_{75\%}$. In our study, the *environmental flows* scenario is very much conservative, as these conditions occur, on average, less than 1% of days over the year. However, while the use of *average flows* protects the environment against current scenarios of pollution, the use of *environmental flows* would help to protect it against future scenarios of pollution in a climate change context. These results show the importance of selecting the appropriate hydrological conditions when proposing the optimal strategy for the removal of microcontaminants. Hydrological characteristics of European rivers vary greatly, and higher costs are expected for basins with low discharge, such as the Mediterranean basins. In addition, Mediterranean rivers will be particularly affected by climate change, as climate projections predict even lower discharges by the end of the century (Pascual et al., 2014).

4.5. Recommendations for decision-makers to upgrade WWTPs for the removal of diclofenac

This study provides an overall goal and a realistic budget to decision-makers. The recommendation for them would be to invest in the development of a model with low uncertainty that would then be used for decision-making. The investment in a research project (e.g., 1 M€ as the total cost of the European Industrial Doctorate – TreatRec) would be paid back in less than one year due to the reduction in the cost of the upgrades when simulating the model with reduced parameter uncertainty. The costs of constructing and operating tertiary treatments are in the order of magnitude of 10 M€·year^{-1}, and hence, any investment to enhance the prediction capabilities of the model will result in enormous savings, even in the short term. We believe that adaptive management is an excellent approach to accommodating for future uncertainties and hydrological scenarios. Hence, we would suggest not upgrading all WWTPs at once but starting with the most relevant ones. Our study demonstrated that 3 WWTPs (Rubi, Terrassa and Sant Feliu) can be prioritized in their investment plans since they are included in every solution regardless of the EQS, uncertainty and hydrological condition. With regards to the trade-off between the EQS and cost, we found that there are no large differences in the number of WWTPs requiring upgrade and the costs between 30 and 50 ng L^{-1}. Hence, a good conservative solution would be to set an EQS of 30 ng L^{-1}, which involves the upgrade of 8 WWTPs and 6.3 M€·year^{-1} for *average flows* and the upgrade of 17 WWTPs and 11.1 M€·year^{-1} for *environmental flows*. Given the power relationship between EQSs and costs, going lower (to 10 ng L^{-1}) would be too precautionary. Going higher (100 ng L^{-1}) might endanger the freshwater ecosystem (Ecotox Centre, 2017).

The model used in this study can be applied to any worldwide catchment as the Matlab code and the specific data of each catchment are well separated. The model uses data that is readily available from Environment and Health Agencies and River Basin Authorities (i.e. river network, flows and velocities, WWTP operational parameters, population connected to the WWTPs, consumption of pharmaceuticals). Moreover, there is no need for user's knowledge on river hydrodynamics to run the model (river flows are imported from other models or from measurements at monitoring stations).

4.6. Limitations of this study

We acknowledge that the final solution adopted by the Catalan Water Agency must be valid for the removal of a number of representative microcontaminants in the Llobregat, not only for diclofenac. Our model is ready to simulate these micro-contaminants, but additional modeling efforts are needed to combine the optimal set of solutions for each microcontaminant (e.g. adjustments in the Matlab code to account for a number of microcontaminants and computational time increase). This study focuses on the influence that the uncertainty in the model parameters - F, k_{WWTP} and k_{river} - has on the estimates of pharmaceutical concentrations in the rivers, and, in turn, on the EQS exceedance, and ultimately, on the cost of the upgrades. Other sources of uncertainty (e.g. 18% of uncertainty in the ozonation costs, Mulder et al., 2015) are not the focus of this study. Finally, source control measures are not considered, and we will address this issue in our next study.

5. Conclusions

The cost of the WWTP upgrades decreases non-linearly (from

10.1 to 4.8 M€·year^{-1} for average flows and from 11.1 to 8.8 M€·year^{-1} for environmental flows) as the EQS increases from 10 to 100 ng/l. Setting 30 ng/l as the EQS for diclofenac would balance costs and ecosystems protection. Our methodology helps river basin authorities find the optimal set of WWTPs that should be upgraded for different EQSs.

Searching for the optimal set of WWTPs that should be upgraded to comply with EQSs of microcontaminants is a catchment-specific problem. Establishing a uniform strategy in Europe for the upgrade of WWTPs seems to be challenging and suboptimal.

Investing in research projects aimed at decreasing model parameter uncertainty leads to enormous savings in the cost of the WWTP upgrades (down to 1.3 M€·year^{-1} for 1.1 M inhabitants), which would have a positive effect on our annual water bill (reductions of up to 4 €·household^{-1}·year^{-1}).

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Appendix A. Supplementary data

Supplementary data related to this article can be found at https://doi.org/10.1016/j.watres.2018.07.002.

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