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Environmental optimization of continuous flow ozonation for urban wastewater reclamation

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Abstract

Wastewater samples from two treatment plants were spiked in the microgram-to-tens-of-microgram per litre range with diuron (herbicide), ibuprofen and diclofenac (anti-inflammatory drugs), sulfamethoxazole and erythromycin (antibiotics), bezafibrate and gemfibrozil (lipid regulators), atenolol (β-blocker), carbamazepine (anti-epileptic), hydrochlorothiazide (diuretic), caffeine (stimulant) and N-acetyl-4-amino-antipiryne, a metabolite of the antipyretic drug dypirone. They were subsequently ozonated in continuous flow using bubble columns. The concentration of all spiking compounds was monitored in the outlet stream. The effects of varying ozone input, expressed as energy per unit volume, and water flow rate, and of using single or double column were studied in relation to the efficiency of ozone usage and the ratio of pollutant depletion. The ozone dosage required to treat both wastewaters with pollutant depletion > 90% was in the 5.5-8.5 mg/L range with ozone efficiencies greater than 80% depending on the type of wastewater and the operating conditions. This represented 100-200 moles of ozone transferred per mole of pollutant removed. Direct and indirect environmental impacts of ozonation were assessed according to Life Cycle Assessment, a technique that helped identify the most effective treatments in terms of potential toxicity reduction, as well as of toxicity reduction per unit mass of greenhouse-gas emissions, which were used as an indicator of environmental efficiency. A trade-off between environmental effectiveness (toxicity reduction) and greenhouse-gas emissions was observed since maximizing toxicity removal led to higher greenhouse-gas emissions, due to the latter's relatively high ozone requirements. Also, there is an environmental trade-off between effectiveness and efficiency. Our results indicate that the choice of the most desirable treatment alternative depends on whether the goal pursued is maximum possible pollutant removal less than maximum but more efficient pollutant removal.

Keywords: Ozone, Life Cycle Assessment, reclaimed wastewater, emergent pollutants, continuous ozonation.

1. Introduction

According to the United Nations, 47% of the world's population will be living in water-stressed areas by 2050 (UNESCO, 2009). In addition, the lack of facilities for treating wastewater before its discharge in water bodies will contribute to a freshwater shortage. In view of this situation, the European Union has developed a common water policy (Directive 2000/60/EC) with the stated aims of improving the quality of water bodies and promoting the protection and sustainable use of the water environment. In particular, the Spanish authorities have declared their intention of adopting measures in order to attain the complete treatment of wastewater effluents from agglomerations of more than 2000 equivalent inhabitants (e.i.) and to increase by a factor of two (up to 463 hm^3/year) the volume of reclaimed wastewater in the near future. The widespread use of reclaimed water may contribute significantly to reducing the pressure on water resources in countries such as Spain and other southern Europe countries, which are considered water-stressed by the European Environment Agency (European Environment Agency, 2005). Among the main reasons why wastewater reuse has not received greater attention to date are the potential effects on human health and concern over the release to the environment of trace compounds such as priority and emerging pollutants, including pharmaceuticals and personal care products (PPCP). It is well-known that these substances escape from conventional activated sludge wastewater treatment plants and thus enter surface water streams (Ternes, 2007; Rosal et al., 2010)

Ozone is a strong oxidant and its potential for the depletion of organic pollutants has been studied extensively over the last few years (Ikehata et al., 2008;

Rosal et al., 2009). Ozonation is an effective process for reclaiming wastewater and could provide resources for use in applications where freshwater quality is not necessary. However, many common water pollutants have low direct ozonation rate constants (Hoigné and Bader, 1983). Therefore, the effective depletion of many refractory compounds relies on the generation of secondary oxidants like hydroxyl radicals, which are produced by so-called advanced oxidation processes (AOP). Ozone-based advanced oxidation processes (O_3-AOP) have proven to be effective for removing priority and PPCP pollutants from wastewater and may be implemented simply by keeping a basic medium (Rosal et al., 2008; Sievers, 2011). Although O_3-AOP have been applied in a number of bench and pilot studies as tertiary treatments for the effluents of wastewater treatment plants (WWTP), the application in full-scale processes is still limited (Schaefer et al., 2009). The economic costs associated with ozone generation and the concern about its impact on the environment are hindering its large-scale use. The generalization of this technology also means that its direct and indirect environmental impacts need to be minimized. In a previous study we evaluated ozonation as a tertiary treatment using a LCA approach and focusing on potential toxicity and greenhouse-gas (GHG) emissions (Muñoz et al. 2009). We showed there that reusing wastewater after ozonation had environmental benefits when compared either with reuse with no tertiary treatment or with a non-reuse scenario. However, one of the major limitations of that study was the use of discontinuous data, whereas large-scale ozonation systems operate in continuous mode.

In this study, we undertook a continuous ozonation process in bubble column in order to determine the most suitable operational conditions for reclaiming wastewater using real WWTP effluents spiked with twelve compounds representative of the pollutants typically found in wastewater. The data allowed classic operating parameters, such as hydraulic retention time and ozone dosage to be optimized. The experimental results were used to assess the environmental effectiveness and efficiency of ozone use based on the life cycle assessment (LCA) methodology.

2. Material and methods

2.1. Materials

The ozonation was carried out in two treated wastewater matrices from West-Alcalá (3000 m^3/h, 374000 e.i., Alcalá de Henares, Madrid, Spain), hereinafter AH, and Alcázar de San Juan (1000 m^3/h, 208200 e.i., Ciudad Real, Spain), hereinafter ASJ. Both WWTPs apply an activated sludge treatment followed by clarification. The main sources of these waters are urban and, to a lesser extent, agricultural and industrial. The main characteristics of both wastewaters are shown in Table 1. The spiking of micropollutants was performed in the microgram-to-tens-of-microgram per liter range using compounds selected from those typically found in wastewater: diuron (herbicide), ibuprofen and diclofenac (anti-inflammatory drugs), sulfamethoxazole and erythromycin (antibiotics), bezafibrate and gemfibrozil (lipid regulators), atenolol (β-blocker), carbamazepine (anti-epileptic), hydrochlorothiazide (diuretic), caffeine (stimulant) and a metabolite of the antipyretic drug dypirone, N-acetyl-4-amino-antipiryne (4-AAA). All these chemicals were purchased at analytical grade (purity > 90%) from Sigma–Aldrich. The reference compounds used as surrogate standard, ^{13}C-phenacetin, and ^{13}C-caffeine, were purchased from Dr. Ehrenstorfer. Stock standard solutions of individual compounds were prepared as concentrates in methanol or water and stored at –20ºC. The solvents used for HPLC were methanol and acetonitrile HPLC grade supplied by Merck. Water used for LC-MS analysis was generated from a Direct-Q™ 5 Ultrapure Water Systems from Millipore with a specific resistance of 18.2 MΩ·cm. Commercial cartridges packed with Oasis™ HLB (200 mg, 6 cc) were purchased from Waters. Formic acid (purity, 98 %) was obtained from Fluka.

Table 1. Main wastewater parameters

Parameter	AH	ASJ
pH	7.5	8.4
Total suspended solids (mg/L)	5.7	17.4
Turbidity (NTU)	4	10
Conductivity (μS/cm)	820	2090
Chemical oxygen demand (COD, mg/L)	15.3	17.8
Total organic carbon (TOC, mg/L)	6.0	8.1
Alkalinity (mg/L $CaCO_3$)	198	451
Wastewater matrix (μg/L)		
4-AAA	4.49	11.30
Atenolol	1.03	0.90
Bezafibrate	0.13	0.40
Caffeine	1.18	0.98
Carbamazepine	0.12	0.10
Diclofenac	0.22	0.10
Diuron	0.14	0.09
Erythromycin	0.33	0.10
Gemfibrozil	0.85	0.01
Hydrochlorothiazyde	1.18	2.20
Ibuprofen	0.14	0.10
Sulfamethoxazole	0.23	0.10
Concentrations after spiking (μg/L)		
Diclofenac	13.5	52.0
Ibuprofen	1.5	13.0
Bezafibrate	31.0	16.3
Gemfibrozil	2.7	7.4
Hydrochlorothiazide	9.8	13.6
Atenolol	37.2	27.8
Caffeine	18.4	13.0
Erythromycin	0.9	0.7
Sulfamethoxazole	28.2	16.8
Carbamazepine	19.1	14.2
4-AAA	54.5	92.2
Diuron	0.2	0.4
Total spiking concentration (nM)	832	1035
TOC after spiking (mg/L)	6.1	8.3

2.2. Analyses

Aliquots of treated sample (500 mL) were collected by using pre-rinsed amber glass bottles, stored in the dark at 4ºC until analysis and extracted within 48 h in all cases. Before extraction each sample was filtered through a 0.7 μm glass fibre filter (Teknokroma, Barcelona, Spain). A solid phase extraction (SPE) procedure was applied to the samples using commercial OasisTM HLB (divinylbenzene/N-vinylpyrrolidone copolymer) cartridges (200 mg, 6 cc) from Waters (Mildford, MA, USA). An automated sample processor ASPEC XL fitted with an 817 switching valve and an external 306 LC pump from Gilson (Villiers-le-Bel, France) was used for this purpose. The Oasis HLB cartridges were preconditioned with 6 ml of MeOH and 5 mL of deionized water HPLC-grade (pH adjusted to 8 with HCl 2N or NH_4OH 20%) at a flow rate of 1 mL/min. After the conditioning step, aliquots of 400 mL of sample (pH adjusted to 8) were loaded into the cartridge. Samples were previously spiked with 10 μL of 10 mg/L solution of the surrogate standards, ^{13}C-phenacetin and ^{13}C-caffeine. Samples were passed through the cartridges at a flow rate of 10 mL/min and then, rinsed with 5 mL of deionized water prior to elution. After that, the cartridges were dried by nitrogen stream for approximately 5 min to remove excess of water and, finally, the analytes retained were eluted with 2 x 4 ml of MeOH at 1 ml/min. The extracts were evaporated until almost dry using a Turbo-Vap from Zymark (Hopkinton, Massachusetts), with the water temperature at 35ºC. The samples were then reconstituted with 1mL of acrilonitrile:water, 10:90 (v/v) before being filtered directly into an analysis vial using a 0.45 μm PTEF syringe filter (Millipore, USA).

Each extract was analyzed by high-pressure liquid chromatography-tandem mass spectrometry (HPLC-MS/MS) using a 3200 QTRAP triple quadrupole-ion trap mass spectrometer equipped with a turbo ion spray source (AB Sciex Instruments, Foster City, CA). Analytes were determined by ESI-MS/MS either in positive or negative mode by selected reaction monitoring (SRM). All parameters and operating conditions for the analysis are described in detail elsewhere (Martínez Bueno et al., 2007). The following analytes were determined in positive mode: atenolol, caffeine, 4.-AAA, erythromycin, sulfamethoxazole, and carbamazepine. For their part, diclofenac, bezafibrate, gemfibrozil, hydrochlorothiazide, diuron and ibuprofen, were analyzed in negative mode. The separation of the analytes was performed using an HPLC (series 1100, Agilent Technologies, Palo Alto, CA) equipped with a C-18 analytical column, 250 mm x 3.0 mm i.d and 5 μm (ZORBAX SB, Agilent Technologies). For the analysis in positive mode, the compounds were analyzed using acrilonitrile and MiliQ-water with 0.1% formic acid, while acrilonitrile and MiliQ-water were the mobile phases used for analysis in negative mode. The volume injected was 20 μL in both modes. Confirmation of each compound was performed by means of two SRM transitions at the correct retention time and by monitoring the SRM ratio, in accordance with EU guidelines for LC–MS/MS analysis (Commission Decision, 2002/657/EC). An additional experiment was necessary in order to carry out the correct identification of the drug ibuprofen, for which more structural information was required. In this case, triple quadrupole-linear ion trap system (QqQLIT) operated using enhanced product ion (EPI) mode, in the same injection. The confirmation criterion applied for their identification in water samples was the presence of the characteristic SRM transition at the correct retention time and a good match between the reference spectrum of the library (developed by this research group) and the spectrum obtained in the samples, or by a fit value higher than 70%.

2.3. Operational procedure

The loads of the selected pollutants in raw wastewater were 15.5 μg/L (59.2 nM) and 9.9 μg/L (39.3 nM) for ASJ and AH wastewaters respectively. The final concentration reached in spiked wastewater amounted to some 20 times (17.4 for ASJ and 21.2 for AH) the initial molar concentration for the 12 compounds spiked. In mass concentrations these were 267 μg/L (ASJ) and 216 μg/L (AH). The data are shown in Table 1. The values correspond to the analyses of wastewater prior to the runs and to experimental uncertainty in the spiking process. As operational variables, we used the energy input per unit volume expressed in kWh/m^3 (which varied from 0.03 to 0.77. This is the first figure in the experimental notation), the use of single or dual column arrangement (C1 and C2 respectively) and the wastewater flowrate, which was either 0.15 or 0.30 L/min (the latter given as 2xF in the notation). The ozonation reactor consisted of two 1.2 L bubble columns with the option of being operated in series. Ozone was obtained from oxygen using a COM-AD-02 Anseros generator, with a flow of 0.39 NL/min. The concentration of ozone dissolved in the liquid was monitored using a Rosemount 499AOZ amperometric analyzer periodically calibrated with the Indigo Colorimetric Method (SM 4500-O3 B). A Data Acquisition unit digitalized the signals from the concentration of dissolved ozone, pH and temperature with a sampling period of 1 s. The concentration of ozone in gas phase was measured with an Anseros Ozomat GM6000 Pro photometer calibrated against potassium iodide. Fig. 1 shows a scheme of the experimental set-up.

2.4. Life Cycle Assessment methodology

The environmental assessment was carried out following similar methods and assumptions to those of Muñoz et al. (2009). The goal of the assessment was to compare the environmental performance of the treatments of spikedAH and ASJ wastewater. The system under study included: the production of oxygen, electricity, and cooling water for ozone production; the transport of oxygen to the treatment site, assuming a distance of 100 km and the emission of trace pollutants either to a river or to agricultural soil. Emissions to a river were considered when assessing aquatic ecotoxicity, whereas

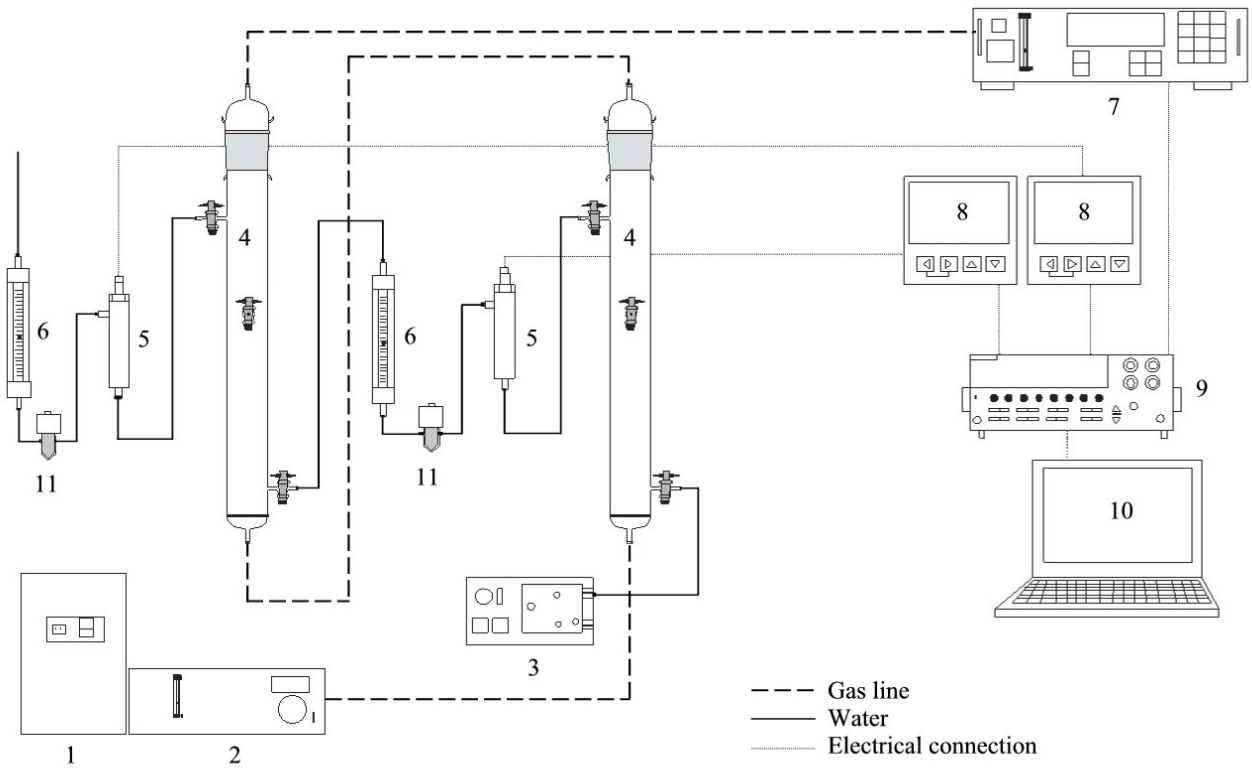

Figure 1. Set-up of the experimental equipment: (1) PSA, (2) Ozone generator, (3) Pump, (4) Bubble column, (5) Ozone sensor, (6) Flowmeter, (7) Gas-phase ozone analyzer, (8) Dissolved ozone analyzer, (9) Data acquisition unit, (10) Computer.

emissions to soil were evaluated in the assessment of terrestrial ecotoxicity and human toxicity.

In the inventory analysis, the ozone input (including ozone consumed and losses) for each treatment was obtained from the experiments, while the environmental impacts per unit mass of ozone were estimated on the basis of the following figures for a full-scale plant according to Muñoz et al. (2009): 15.85 kWh electric consumption, 8.3 kg oxygen and 2 m^3 water. The ecoinvent database v.2.01 was used to model electricity (Dones et al. 2007) and oxygen production (Althaus et al. 2007), whereas for cooling water seawater desalination data from Muñoz and Rodríguez (2008) were used. The impact assessment phase included the following impact categories: (1) aquatic ecotoxicity potential, (2) terrestrial ecotoxicity potential, (3) human toxicity potential and (4) greenhouse-gas (GHG) emissions. Toxicity-related impact categories were assessed with two characterization models, USES-LCA (Huijbregts et al. 2000) and EDIP'97 (Hauschild and Wenzel, 1998), whereas GHG were assessed using the Global Warming Potentials for 100 years according to the Intergovernmental Panel for Climate Change (Forster et al. 2007). The environmental performance of ozone treatments with regard to toxicity reduction was assessed by the following two criteria. (1) Effectiveness, defined as the ability of each treatment to reduce potential toxicity with a life cycle perspective. Effectiveness was assessed per unit volume of wastewater treated, as the balance between the potential toxicity eliminated from the effluent and the potential toxicity produced by upstream activities, such as energy and production of chemicals. (2) Efficiency, as the ratio of the potential toxicity reduction to environmental impact produced. The environmental impact of the treatments was represented by means of their associated GHG emissions.

3. Results and discussion

3.1 Ozone transfer and pollutant depletion

The efficiency of ozone usage, *E*, was calculated from the fraction of ozone lost in the outlet gas using Eq. 1. The amount of ozone effectively transferred per unit volume of the liquid phase was referred to as dosage, *D* (in mg/L or mmol/L):

$$E = \frac{\left(c_{O_3}\right)_{g,in} - \left(c_{O_3}\right)_{g,out}}{\left(c_{O_3}\right)_{g,in}} \qquad [1]$$

$$D = F_G \frac{\left(c_{O_3}\right)_{g,in} - \left(c_{O_3}\right)_{g,out}}{F_L} \qquad [2]$$

Fig. 2 shows the efficiency of ozone usage as a function of the ratio of pollutant depletion expressed in mass concentration units and referring to the twelve spiking compounds. The plot shows the different scenarios assessed, including the use of one single column (C1) or

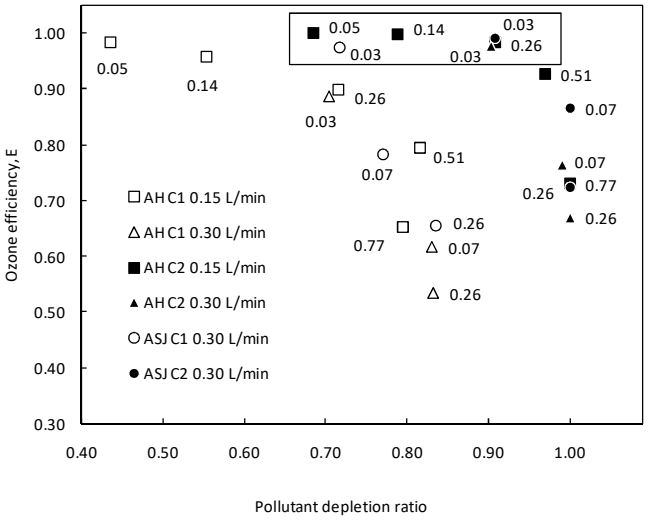

Figure 2. Ozone usage efficiency as a function of pollutant depletion ratio. The labels represent the energy input expressed in kWh/m^3.

two in series (C2). The depletion of pollutants ranged from roughly 50% for the ozonation of AH wastewater in one single column using the highest water flowrate to almost 100% using high concentration of ozone in the gas phase. For wastewater treated in two columns, the pollutant depletion ratio reached up to 98% with ozone efficiency above 90%. By doubling the energy per unit volume of treated water, i.e. by increasing the concentration of ozone in the gas phase, pollutant depletion increased up to almost 100%, but ozone efficiency dropped to 80%. It was possible to obtain a similar pollutant depletion ratio by using one single column, but in this case ozone efficiency was typically 20% lower. The labels indicate the energy used for ozone generation expressed in kWh/m^3. It is apparent that for similar energy inputs the pollutant depletion obtained by two columns was considerably higher than that obtained by one single column, and this is obviously related to ozone usage efficiency. The rectangle in the upper part of the plot shows the best ozonation scenarios, which were those with energy input in the 0.03–0.26 kW-h/m^3 range, ozone efficiency in the 90–100% range and joint pollutant depletion in the 67–98% range. The ozone doses transferred to water, *D*, were in the 5.5-8.5 mg/L range depending on the reactor arrangement and the operational conditions used. This represents 100-200 moles of ozone transferred per mole of pollutant removed in spiked wastewater and, therefore, this figure would be roughly valid for other concentrations or pollutants and for non-spiked wastewater. It is interesting to note that the removal of organic carbon (degree of mineralization) expressed as total organic carbon was limited and in any case over 15%. This figure is comparable to other alkaline ozonation processes, as previously described (Rosal et al., 2008).

3.2. Life Cycle Assessment results

Fig. 3 shows the results of the LCA comparison in toxicity-related impact categories, using a relative scale. The potential toxicity of the effluent prior to ozonation is taken as the reference, with the alternative treatments shown as a percentage of that initial effluent. For each impact category and treatment, the results according to the USES-LCA and EDIP'97 characterization models are given. The general pattern observed in these graphs is that potential toxicity is reduced by applying any of the ozonation treatments tested. The reduction was generally higher in terms of ecotoxicity (aquatic and terrestrial) rather than in terms of potential effects on humans. In human toxicity, the pollutant which made the greatest contribution was gemfibrozil followed by hydrochlorothiazide. Accordingly, the higher human ecotoxicity potential displayed by treated ASJ wastewaters was due to a higher concentration of both compounds. Both gemfibrozil and hydrochlorothiazide are compounds that react with molecular ozone at a relatively high rate. The second order rate constant for the direct ozonation of hydrochlorothiazide at pH 9 is 16400 M^{-1} s^{-1} (Real et al., 2010). For gemfibrozil, we calculated a rate constant of 42900 ± 5200 M^{-1} s^{-1}, obtained using competition kinetics with isoproturon in batch ozonation runs. In these conditions, higher pH values favour the formation of hydroxyl radicals due to the reaction with hydroxide anion (Tomiyasu et al. 1985). At the same time ozone is withdrawn from solution, which is detrimental for the depletion of compounds reacting at a high rate with molecular ozone. For ASJ wastewater, only 0.26-C1 and 0.26-C2 treatments, those with the higher ozone concentration in the gas phase, effectively removed gemfibrozil.

The graphs in Fig. 3 show the total reduction of potential toxicity, including that from the effluent pollutants but also from upstream activities (production of electricity, chemicals, etc.). Although the contribution of the latter is not shown explicitly in the graphs, these upstream activities have a variable contribution, depending on the model used. According to USES-LCA, the contribution of utilities is only relevant in human toxicity, whereas according to EDIP'97 utilities are only relevant in aquatic ecotoxicity. As for the overall life-cycle effectiveness of the tested treatments in reducing toxicity, it can be seen that there is no straightforward answer. In aquatic ecotoxicity, USES-LCA promotes 0.26-C1 and 0.26-C2 in ASJ and 0.77-C1/C2 in AH but according to EDIP'97 the potential toxicity of upstream activities makes these treatments less preferable than others using lower ozone concentration in the gas phase. However, in terrestrial ecotoxicity both models coincide in identifying the aforementioned treatments as the most effective. Finally, in human toxicity both models and especially EDIP'97 identify again 0.26-C1 and 0.26-C2 as the most effective for ASJ and 0.77-C1/C2 for AH wastewater. For terrestrial and human toxicity USES-LCA showed a lower discriminating power again due to the weight of upstream activities. The toxicity characterization models used, USES-LCA and EDIP'97, were useful when to identifying the general pattern, i.e., an overall ecotoxicity and human toxicity reduction when ozone is applied to

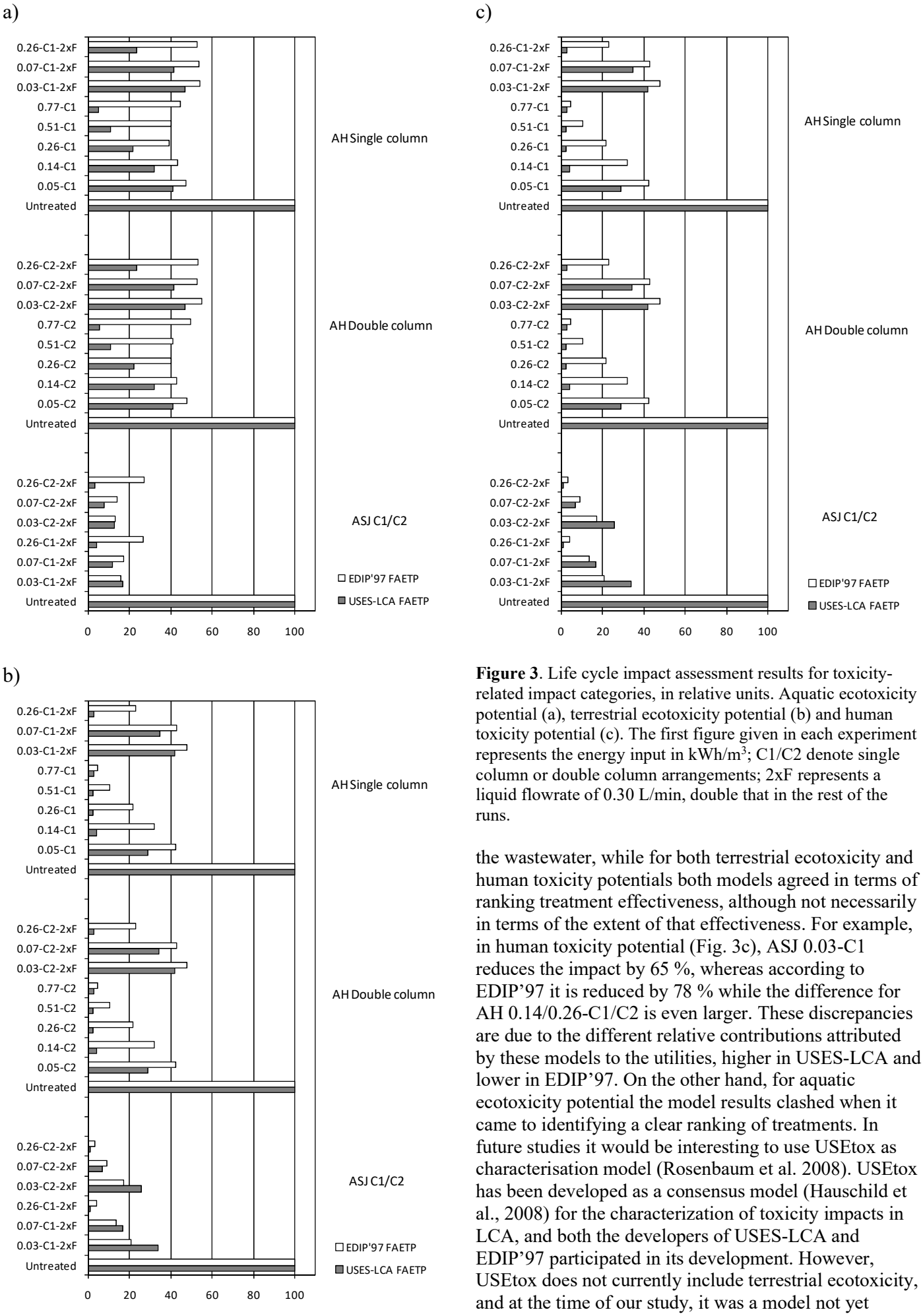

Figure 3. Life cycle impact assessment results for toxicity-related impact categories, in relative units. Aquatic ecotoxicity potential (a), terrestrial ecotoxicity potential (b) and human toxicity potential (c). The first figure given in each experiment represents the energy input in kWh/m^3; C1/C2 denote single column or double column arrangements; 2xF represents a liquid flowrate of 0.30 L/min, double that in the rest of the runs.

the wastewater, while for both terrestrial ecotoxicity and human toxicity potentials both models agreed in terms of ranking treatment effectiveness, although not necessarily in terms of the extent of that effectiveness. For example, in human toxicity potential (Fig. 3c), ASJ 0.03-C1 reduces the impact by 65 %, whereas according to EDIP'97 it is reduced by 78 % while the difference for AH 0.14/0.26-C1/C2 is even larger. These discrepancies are due to the different relative contributions attributed by these models to the utilities, higher in USES-LCA and lower in EDIP'97. On the other hand, for aquatic ecotoxicity potential the model results clashed when it came to identifying a clear ranking of treatments. In future studies it would be interesting to use USEtox as characterisation model (Rosenbaum et al. 2008). USEtox has been developed as a consensus model (Hauschild et al., 2008) for the characterization of toxicity impacts in LCA, and both the developers of USES-LCA and EDIP'97 participated in its development. However, USEtox does not currently include terrestrial ecotoxicity, and at the time of our study, it was a model not yet

implemented in LCA software and thus difficult to apply in practice.

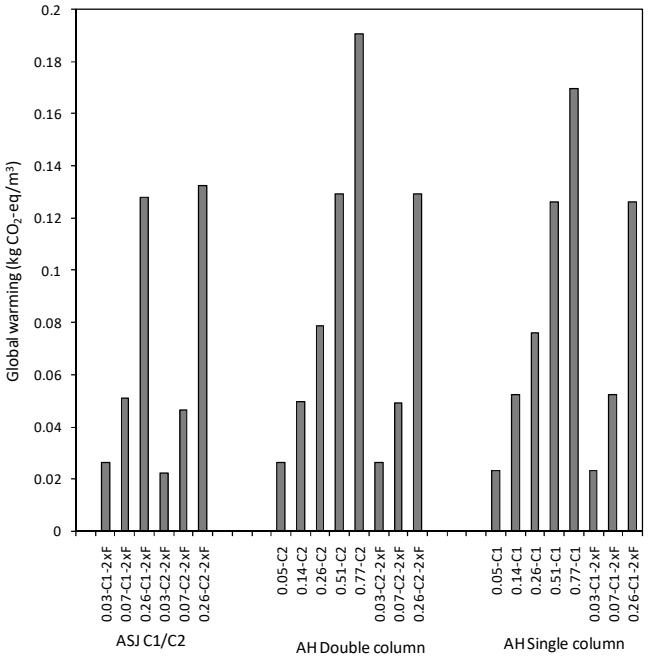

Figure 4. Life cycle impact assessment results for greenhouse-gas emissions per unit volume treated.

Fig. 4 shows the GHG emissions per unit volume effluent treated and for each treatment tested. Emissions range from 0.02 to 0.20 kg CO_2-eq./m^3. These emissions are mainly related to electricity and oxygen production, and are proportional to the ozone produced per unit volume of the liquid phase. If we compare GHG emissions and potential toxicity reduction we observe that there is a clear link between pollutant removal efficiency and GHG emissions: those treatments, such as ASJ 0.26-C1 and ASJ 0.26-C2, that achieve the highest removal of micropollutants, also entail the highest GHG emissions. When comparing the degree of mineralization and GHG emissions, we found a direct relationship for lower energy input that reached a plateau for the highest concentrations of ozone in the gas phase. For the whole set of experiments, the removal ratio was in the 0.04-0.12 kg TOC removed/kg CO_2-eq. range. The results of the efficiency assessment are summarized in Table 2 which shows the effect on aquatic, terrestrial and human ecotoxicity potential in relation to the GHG emissions (expressed in kg DCB-eq/kgCO2-eq, where DCB, which stands for 1,4-dichlorobenzene is the reference substance for toxicity). The ratio of potential toxicity reduction to GHG emissions can be calculated for the three toxicity-related impact categories, and for the two characterization models. In order to interpret this table, numbers in the same row must be compared with one another; the higher the number, the higher the efficiency (higher toxicity removal per kg CO_2-eq emitted). It will be seen that lower ozone generations (AH runs using 0.05 kWh/m^3 and ASJ runs 0.03-C1/C2) allow the highest values for the ratio to be achieved for all impact categories (aquatic, terrestrial, human) and for both models. Therefore, these treatments can be considered the most efficient from a GHG emission perspective. On the other hand, high intensity treatments such as AH 0.77-C1/C2 obtain the lowest ratio values and can be considered as the least efficient.

All the tested treatments were effective in regard of potential toxicity. Applying ozone in continuous mode significantly reduces the pollutant content of the effluent, and this is not counterbalanced by the emissions of pollutants associated with producing electricity or manufacturing and transporting oxygen. However, the higher the ozone input to the columns, the higher the GHG emissions. Therefore a trade-off between toxicity and GHG emissions occurs. These results show that in order to choose the most convenient treatment option, the goal of the treatment must be established first. If we aim to reduce potential toxicity as much as possible (effectiveness), then it is preferable to use high ozone inputs as they lead to more effective pollution removal and to use a double column arrangement rather than a single bubble column (points aligned vertically at near 100% removal in Fig. 2). This stands true even after taking into account the upstream impacts of utilities. On the other hand, these treatments are the least efficient as they entail the highest GHG emissions per unit volume of effluent treated and the lowest toxicity removal per kg CO_2-eq. emitted. If we aim to minimize the toxicity- to-GHG trade-off, then the low intensity treatments are the most appropriate regardless of whether single or double column arrangements are used. For these conditions in ASJ wastewater the use of C2 is preferable, while for AH C1 yielded better results, but the differences were minimal (Table 2). The absolute GHG emissions, estimated in the 0.02 to 0.20 kg CO_2-eq/m^3 range are significantly lower to those estimated by Muñoz et al. (2009) of 0.27 kg CO_2-eq/m^3. This figure was derived on the bases of discontinuous bench-scale experiments and a series of assumptions on up-scaling. It is important to stress that even though the present experiments have also been carried out at laboratory scale, they are closer to a full-scale system, since a reactor working in continuous mode was used. GHG emissions of ozonation were compared with those of other advanced wastewater treatments. Meneses et al. (2010) assessed an advanced disinfection treatment consisting of chlorination plus ultraviolet (UV) irradiation and compared its impact to that of ozonation as per Muñoz et al. (2009). The disinfection treatment entailed emissions of 0.2 kg CO_2-eq/m^3, about 50% lower than those produced by ozone. In the present work, the emissions per unit volume, assessed using a continuous system, are up to one order of magnitude lower than those of chlorination followed by disinfection as per Meneses et al. (2010). It should be noted that Meneses et al. (2010) used data from a full scale existing WWTP that was applying a disinfection treatment, whereas the present study is based on laboratory experiments with spiked wastewater, as a consequence of which these studies can only be compared to a degree.

Table 2. Assessment of environmental efficiency for ozone treatments: ratio of potential toxicity reduction to GHG emissions. (Notes: Toxicity in USES-LCA is measured in kg of a reference substance, namely 1,4-dichlorobenzene (DCB). Toxicity in EDIP'97 is measured as the volume of environmental compartment (water or soil) needed to dilute emissions to a no effect concentration. For the notation of the experiments, see the caption of Fig. 3.

Treatment									
AH – Single Column (C1)		0.05	0.14	0.26	0.51	0.77	0.03-2xF	0.07-2xF	0.07-2xF
USES-LCA	Aquatic ecotoxicity potential (kg DCB-eq/kgCO_2-eq)	167	87	68	47	37	151	75	40
	Terrestrial ecotoxicity potential (kg DCB-eq/kgCO_2-eq)	1.1	0.6	0.4	0.3	0.2	0.9	0.4	0.3
	Human toxicity potential (kg DCB-eq/kgCO_2-eq)	3.7	1.7	1.3	0.8	0.6	4.1	1.6	0.8
EDIP'97	Aquatic ecotoxicity potential (m^3 water/kgCO_2-eq)	23358	11172	8256	4889	3370	20225	9167	3880
	Terrestrial ecotoxicity potential (m^3 soil/kgCO_2-eq)	48411	25423	20139	13888	10986	43798	21372	11888
	Human toxicity potential (m^3 soil/kgCO_2-eq)	28.7	12.9	8.9	5.4	4.0	28.5	12.7	5.3
AH – Double Column (C2)		0.05	0.14	0.26	0.51	0.77	0.03-2xF	0.07-2xF	0.26-2xF
USES-LCA	Aquatic ecotoxicity potential (kg DCB-eq/kgCO_2-eq)	149	91	66	46	33	133	79	39
	Terrestrial ecotoxicity potential (kg DCB-eq/kgCO_2-eq)	0.9	0.7	0.4	0.3	0.2	0.8	0.5	0.3
	Human toxicity potential (kg DCB-eq/kgCO_2-eq)	3.3	1.8	1.2	0.7	0.5	3.6	1.7	0.7
EDIP'97	Aquatic ecotoxicity potential (m^3 water/kgCO_2-eq)	20437	11880	7854	4699	2730	17496	9895	3716
	Terrestrial ecotoxicity potential (m^3 soil/kgCO_2-eq)	42928	26745	19382	13529	9777	38524	22713	11581
	Human toxicity potential (m^3 soil/kgCO_2-eq)	25.4	13.5	8.6	5.3	3.6	25.0	13.5	5.2
ASJ- Single / Double Column		0.03-C1-2xF	0.07-C1-2xF	0.26-C1-2xF	0.03-C2-2xF	0.07-C2-2xF	0.26-C2-2xF		
USES-LCA	Aquatic ecotoxicity potential (kg DCB-eq/kgCO_2-eq)	164	90	39	201	102	38		
	Terrestrial ecotoxicity potential (kg DCB-eq/kgCO_2-eq)	1.5	1.0	0.5	1.9	1.2	0.4		
	Human toxicity potential (kg DCB-eq/kgCO_2-eq)	2.9	1.8	1.6	6.7	3.4	1.6		
EDIP'97	Aquatic ecotoxicity potential (m^3 water/kgCO_2-eq)	40594	20705	7262	49288	23460	6988		
	Terrestrial ecotoxicity potential (m^3 soil/kgCO_2-eq)	38641	21866	9632	47362	25053	9392		
	Human toxicity potential (m^3 soil/kgCO_2-eq)	19	12	12	52	26	12		

Conclusions

The depletion of micropollutants with ozone increased with rising energy input, which is directly related to the concentration of ozone in the gas phase. Beyond certain energy intensity, expressed in kWh/m^3, further increases in pollutant removal are accompanied by a sharp decrease in ozone usage efficiency. For the best ozonation scenarios, the energy input was in the 0.03–0.26 kWh/m^3 range with ozone efficiencies in the 90–100% range and joint pollutant depletion in the 67–98% range. In all cases, the arrangement of two columns in series yielded better results, particularly for the efficiency of ozone usage, which was typically 20% higher. The ozone doses transferred to wastewater were in the 5.5-8.5

mg/L range depending on the reactor arrangement and the operating conditions used; this represented 100-200 moles of ozone transferred per mole of pollutant removed.

LCA was applied to compare the environmental performance of different alternatives for using ozone to remove micropollutants into a wastewater reuse scheme. Based on laboratory-scale continuous flow experiments performed in bubble column, it was found that the most effective ozone treatments for the removal of potential toxicity also entail higher GHG emissions due to their relatively high ozone requirements. The ozone treatments entailing the lowest ozone input and therefore lowest GHG emissions were the most efficient among those tested since they maximized the potential toxicity removed per unit mass GHG emitted. Therefore, there is a trade-off between two different environmental impacts: toxicity and GHG emissions. The choice of the most desirable treatment alternative will depend on whether the goal of the treatment is to maximize toxicity removal or achieve a given reduction target with the minimal energy use and GHG emissions.

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Supplementary Information

Environmental optimization of continuous flow ozonation for urban wastewater reclamation

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Table 1. Contributions to toxicity impact categories modelled with USES-LCA, from pollutants in effluents with and without treatment of Alcázar de San Juan wastewater. Contributions from energy and chemicals for treatments not included here. Scores per m^3 wastewater.

Pollutants	Impact scores FAETP USES-LCA emissions to water (kg 1,4-DCB/m3 wastewater)						
	Untreated	0.03-C1-2xF	0.07-C1-2xF	0.26-C1-2xF	0.03-C2-2xF	0.07-C2-2xF	0.26-C2-2xF
Atenolol	5.2E-04	2.8E-04	2.5E-04	4.5E-05	2.4E-04	2.3E-04	5.6E-06
Bezafibrate	1.5E+00	2.9E-01	2.8E-01	1.6E-01	2.8E-01	2.7E-01	1.3E-01
Caffeine	5.5E-04	1.9E-04	1.6E-04	9.3E-05	1.7E-04	1.3E-04	2.5E-05
Carbamazepine	3.3E-05	1.6E-05	8.3E-06	0.0E+00	1.4E-05	5.6E-06	0.0E+00
Diclofenac	8.2E-03	2.8E-03	1.4E-03	1.6E-05	2.4E-03	5.9E-04	1.6E-05
Diuron	3.8E-03	9.4E-04	9.4E-04	0.0E+00	9.4E-04	9.4E-04	0.0E+00
Erythromycin	1.7E-01	2.5E-02	0.0E+00	0.0E+00	0.0E+00	0.0E+00	0.0E+00
Gemfibrozil	1.3E-01	9.3E-02	8.2E-02	0.0E+00	3.6E-02	3.2E-02	0.0E+00
Hydrochlorothiazide	3.3E-05	1.1E-05	1.0E-05	4.9E-07	1.1E-05	8.1E-06	0.0E+00
Ibuprofen	2.0E+00	0.0E+00	0.0E+00	0.0E+00	0.0E+00	0.0E+00	0.0E+00
N-acetyl-4-amino-antipiryne (4-AAA)	1.3E+00	4.3E-01	2.1E-01	0.0E+00	3.3E-01	8.2E-02	0.0E+00
Sulfamethoxazole	4.7E-02	1.0E-02	2.2E-03	2.8E-04	5.3E-03	5.5E-04	0.0E+00
	5.2E+00	**8.5E-01**	**5.8E-01**	**1.6E-01**	**6.6E-01**	**3.9E-01**	**1.3E-01**

Table 1. Cont.

Pollutants	Impact scores TETP USES-LCA emissions to soil (kg 1,4-DCB/m3 wastewater)						
	Untreated	0.03-C1-2xF	0.07-C1-2xF	0.26-C1-2xF	0.03-C2-2xF	0.07-C2-2xF	0.26-C2-2xF
Atenolol	2.1E-05	1.1E-05	1.0E-05	1.8E-06	9.8E-06	9.4E-06	2.2E-07
Bezafibrate	3.7E-04	6.9E-05	6.7E-05	3.8E-05	6.7E-05	6.5E-05	3.1E-05
Caffeine	1.9E-05	6.7E-06	5.6E-06	3.3E-06	5.9E-06	4.4E-06	8.9E-07
Carbamazepine	1.3E-07	6.2E-08	3.2E-08	0.0E+00	5.4E-08	2.1E-08	0.0E+00
Diclofenac	1.1E-06	3.8E-07	1.9E-07	2.2E-09	3.3E-07	8.0E-08	2.2E-09
Diuron	9.1E-06	2.3E-06	2.3E-06	0.0E+00	2.3E-06	2.3E-06	0.0E+00
Erythromycin	4.5E-04	6.4E-05	0.0E+00	0.0E+00	0.0E+00	0.0E+00	0.0E+00
Gemfibrozil	1.4E-05	9.6E-06	8.5E-06	0.0E+00	3.7E-06	3.3E-06	0.0E+00
Hydrochlorothiazide	1.4E-06	4.7E-07	4.2E-07	2.1E-08	4.5E-07	3.4E-07	0.0E+00
Ibuprofen	5.2E-04	0.0E+00	0.0E+00	0.0E+00	0.0E+00	0.0E+00	0.0E+00
N-acetyl-4-amino-antipiryne (4-AAA)	5.6E-02	1.9E-02	9.3E-03	0.0E+00	1.4E-02	3.6E-03	0.0E+00
Sulfamethoxazole	1.2E-03	2.5E-04	5.6E-05	7.0E-06	1.3E-04	1.4E-05	0.0E+00
	5.8E-02	**1.9E-02**	**9.4E-03**	**5.0E-05**	**1.5E-02**	**3.7E-03**	**3.2E-05**

Table 1. Cont.

Pollutants	Impact scores HTP USES-LCA emissions to soil (kg 1,4-DCB/m3 wastewater)						
	Untreated	0.03-C1-2xF	0.07-C1-2xF	0.26-C1-2xF	0.03-C2-2xF	0.07-C2-2xF	0.26-C2-2xF
Atenolol	1.7E-03	9.3E-04	8.5E-04	1.5E-04	8.2E-04	7.8E-04	1.9E-05
Bezafibrate	6.8E-05	1.3E-05	1.3E-05	7.1E-06	1.3E-05	1.2E-05	5.9E-06
Caffeine	4.9E-04	1.7E-04	1.4E-04	8.4E-05	1.5E-04	1.1E-04	2.3E-05
Carbamazepine	4.2E-04	2.1E-04	1.1E-04	0.0E+00	1.8E-04	7.1E-05	0.0E+00
Diclofenac	5.6E-05	1.9E-05	9.4E-06	1.1E-07	1.7E-05	4.0E-06	1.1E-07
Diuron	5.1E-04	1.3E-04	1.3E-04	0.0E+00	1.3E-04	1.3E-04	0.0E+00
Erythromycin	4.3E-05	6.2E-06	0.0E+00	0.0E+00	0.0E+00	0.0E+00	0.0E+00
Gemfibrozil	1.7E-01	1.2E-01	1.1E-01	0.0E+00	4.7E-02	4.2E-02	0.0E+00
Hydrochlorothiazide	3.4E-02	1.2E-02	1.0E-02	5.0E-04	1.1E-02	8.3E-03	0.0E+00
Ibuprofen	2.0E-05	0.0E+00	0.0E+00	0.0E+00	0.0E+00	0.0E+00	0.0E+00
N-acetyl-4-amino-antipiryne (4-AAA)	0.0E+00	0.0E+00	0.0E+00	0.0E+00	0.0E+00	0.0E+00	0.0E+00
Sulfamethoxazole	1.3E-04	2.7E-05	6.0E-06	7.5E-07	1.4E-05	1.5E-06	0.0E+00
	2.1E-01	**1.3E-01**	**1.2E-01**	**7.4E-04**	**5.9E-02**	**5.1E-02**	**4.8E-05**

Table 2. Contributions to toxicity impact categories modelled with EDIP'97, from pollutants in effluents with and without treatment of Alcázar de San Juan wastewater. Contributions from energy and chemicals for treatments not included here. Scores per m^3 wastewater.

Pollutants	Impact scores FAETP EDIP'97 emissions to water (m3 water/m3 wastewater)						
	Untreated	0.03-C1-2xF	0.07-C1-2xF	0.26-C1-2xF	0.03-C2-2xF	0.07-C2-2xF	0.26-C2-2xF
Atenolol	4.4E-02	2.4E-02	2.2E-02	3.8E-03	2.1E-02	2.0E-02	4.8E-04
Bezafibrate	2.3E+02	4.4E+01	4.2E+01	2.4E+01	4.2E+01	4.1E+01	2.0E+01
Caffeine	1.9E-01	6.6E-02	5.5E-02	3.2E-02	5.8E-02	4.4E-02	8.8E-03
Carbamazepine	5.7E+00	2.8E+00	1.4E+00	0.0E+00	2.4E+00	9.6E-01	0.0E+00
Diclofenac	1.8E+00	6.2E-01	3.1E-01	3.5E-03	5.4E-01	1.3E-01	3.5E-03
Diuron	2.0E+00	5.0E-01	5.0E-01	0.0E+00	5.0E-01	5.0E-01	0.0E+00
Erythromycin	1.4E+01	2.0E+00	0.0E+00	0.0E+00	0.0E+00	0.0E+00	0.0E+00
Gemfibrozil	1.8E+01	1.3E+01	1.1E+01	0.0E+00	4.8E+00	4.4E+00	0.0E+00
Hydrochlorothiazide	4.3E-03	1.5E-03	1.3E-03	6.3E-05	1.4E-03	1.0E-03	0.0E+00
Ibuprofen	7.7E+02	0.0E+00	0.0E+00	0.0E+00	0.0E+00	0.0E+00	0.0E+00
N-acetyl-4-amino-antipiryne (4-AAA)	2.2E+02	7.4E+01	3.6E+01	0.0E+00	5.7E+01	1.4E+01	0.0E+00
Sulfamethoxazole	8.0E+00	1.7E+00	3.8E-01	4.8E-02	9.0E-01	9.5E-02	0.0E+00
	1.3E+03	**1.4E+02**	**9.2E+01**	**2.4E+01**	**1.1E+02**	**6.1E+01**	**2.0E+01**

Table 2. Cont.

Pollutants	Impact scores TETP EDIP'97 emissions to soil (m3 soil/m3 wastewater)						
	Untreated	0.03-C1-2xF	0.07-C1-2xF	0.26-C1-2xF	0.03-C2-2xF	0.07-C2-2xF	0.26-C2-2xF
Atenolol	1.0E-01	5.3E-02	4.9E-02	8.6E-03	4.7E-02	4.5E-02	1.1E-03
Bezafibrate	4.6E+02	8.7E+01	8.4E+01	4.8E+01	8.4E+01	8.1E+01	3.9E+01
Caffeine	4.5E-01	1.5E-01	1.3E-01	7.5E-02	1.4E-01	1.0E-01	2.1E-02
Carbamazepine	6.4E-01	3.2E-01	1.6E-01	0.0E+00	2.8E-01	1.1E-01	0.0E+00
Diclofenac	1.0E+00	3.5E-01	1.7E-01	2.0E-03	3.1E-01	7.3E-02	2.0E-03
Diuron	1.4E-01	3.4E-02	3.4E-02	0.0E+00	3.4E-02	3.4E-02	0.0E+00
Erythromycin	4.1E-01	5.8E-02	0.0E+00	0.0E+00	0.0E+00	0.0E+00	0.0E+00
Gemfibrozil	1.9E+00	1.4E+00	1.2E+00	0.0E+00	5.2E-01	4.7E-01	0.0E+00
Hydrochlorothiazide	1.0E-02	3.4E-03	3.1E-03	1.5E-04	3.3E-03	2.5E-03	0.0E+00
Ibuprofen	2.9E+02	0.0E+00	0.0E+00	0.0E+00	0.0E+00	0.0E+00	0.0E+00
N-acetyl-4-amino-antipiryne (4-AAA)	5.1E+02	1.8E+02	8.6E+01	0.0E+00	1.3E+02	3.3E+01	0.0E+00
Sulfamethoxazole	1.9E+01	4.2E+00	9.3E-01	1.2E-01	2.2E+00	2.3E-01	0.0E+00
	1.3E+03	**2.7E+02**	**1.7E+02**	**4.8E+01**	**2.2E+02**	**1.2E+02**	**3.9E+01**

Table 2. Cont.

Pollutants	Impact scores HTP EDIP'97 emissions to soil (m3 soil/m3 wastewater)						
	Untreated	0.03-C1-2xF	0.07-C1-2xF	0.26-C1-2xF	0.03-C2-2xF	0.07-C2-2xF	0.26-C2-2xF
Atenolol	6.1E-04	3.2E-04	3.0E-04	5.2E-05	2.9E-04	2.7E-04	6.6E-06
Bezafibrate	1.5E-02	2.9E-03	2.8E-03	1.6E-03	2.8E-03	2.7E-03	1.3E-03
Caffeine	3.3E-04	1.2E-04	9.7E-05	5.6E-05	1.0E-04	7.7E-05	1.5E-05
Carbamazepine	1.3E-04	6.5E-05	3.4E-05	0.0E+00	5.7E-05	2.2E-05	0.0E+00
Diclofenac	5.7E-03	2.0E-03	9.6E-04	1.1E-05	1.7E-03	4.1E-04	1.1E-05
Diuron	2.7E-04	6.7E-05	6.7E-05	0.0E+00	6.7E-05	6.7E-05	0.0E+00
Erythromycin	5.8E-06	8.3E-07	0.0E+00	0.0E+00	0.0E+00	0.0E+00	0.0E+00
Gemfibrozil	1.6E+00	1.1E+00	9.7E-01	0.0E+00	4.2E-01	3.8E-01	0.0E+00
Hydrochlorothiazide	1.8E-02	6.2E-03	5.5E-03	2.7E-04	5.9E-03	4.4E-03	0.0E+00
Ibuprofen	3.2E-03	0.0E+00	0.0E+00	0.0E+00	0.0E+00	0.0E+00	0.0E+00
N-acetyl-4-amino-antipiryne (4-AAA)	0.0E+00	0.0E+00	0.0E+00	0.0E+00	0.0E+00	0.0E+00	0.0E+00
Sulfamethoxazole	1.1E-04	2.4E-05	5.4E-06	6.7E-07	1.3E-05	1.3E-06	0.0E+00
	1.6E+00	**1.1E+00**	**9.8E-01**	**2.0E-03**	**4.3E-01**	**3.9E-01**	**1.3E-03**

Table 3. Contributions to toxicity impact categories modelled with USES-LCA, from effluent (pollutants) and utilities (energy and chemicals) of Alcázar de San Juan wastewater. Scores per m^3 wastewater.

FAETP USES-LCA	Untreated	0.03-C1-2xF	0.07-C1-2xF	0.26-C1-2xF	0.03-C2-2xF	0.07-C2-2xF	0.26-C2-2xF
Utilities	0.0E+00	9.1E-03	1.8E-02	4.4E-02	7.8E-03	1.6E-02	4.6E-02
Effluent	5.2E+00	8.5E-01	5.8E-01	1.6E-01	6.6E-01	3.9E-01	1.3E-01
Total	5.2E+00	8.6E-01	6.0E-01	2.0E-01	6.6E-01	4.0E-01	1.8E-01
TETP USES-LCA	Untreated	0.03-C1-2xF	0.07-C1-2xF	0.26-C1-2xF	0.03-C2-2xF	0.07-C2-2xF	0.26-C2-2xF
Utilities	0.0E+00	1.3E-04	2.5E-04	6.3E-04	1.1E-04	2.3E-04	6.5E-04
Effluent	5.8E-02	1.9E-02	9.4E-03	5.0E-05	1.5E-02	3.7E-03	3.2E-05
Total	5.8E-02	2.0E-02	9.7E-03	6.8E-04	1.5E-02	3.9E-03	6.8E-04
HTP USES-LCA	Untreated	0.03-C1-2xF	0.07-C1-2xF	0.26-C1-2xF	0.03-C2-2xF	0.07-C2-2xF	0.26-C2-2xF
Utilities	0.0E+00	1.1E-02	2.1E-02	5.3E-02	9.4E-03	1.9E-02	5.5E-02
Effluent	2.1E-01	1.3E-01	1.2E-01	7.4E-04	5.9E-02	5.1E-02	4.8E-05
Total	2.1E-01	1.5E-01	1.4E-01	5.4E-02	6.8E-02	7.1E-02	5.5E-02

Table 4. Contributions to toxicity impact categories modelled with USES-LCA, from effluent (pollutants) and utilities (energy and chemicals) of Alcázar de San Juan wastewater. Scores per m^3 wastewater.

FAETP EDIP'97	Untreated	0.03-C1-2xF	0.07-C1-2xF	0.26-C1-2xF	0.03-C2-2xF	0.07-C2-2xF	0.26-C2-2xF
Utilities	0.0E+00	6.4E+01	1.2E+02	3.1E+02	5.5E+01	1.1E+02	3.2E+02
Effluent	1.3E+03	1.4E+02	9.2E+01	2.4E+01	1.1E+02	6.1E+01	2.0E+01
total	1.3E+03	2.0E+02	2.2E+02	3.4E+02	1.6E+02	1.8E+02	3.4E+02
TETP EDIP'97	Untreated	0.03-C1-2xF	0.07-C1-2xF	0.26-C1-2xF	0.03-C2-2xF	0.07-C2-2xF	0.26-C2-2xF
Utilities	0.0E+00	5.0E-01	9.7E-01	2.4E+00	4.3E-01	8.9E-01	2.5E+00
Effluent	1.3E+03	2.7E+02	1.7E+02	4.8E+01	2.2E+02	1.2E+02	3.9E+01
total	1.3E+03	2.7E+02	1.7E+02	5.0E+01	2.2E+02	1.2E+02	4.2E+01
HTP EDIP'97	Untreated	0.03-C1-2xF	0.07-C1-2xF	0.26-C1-2xF	0.03-C2-2xF	0.07-C2-2xF	0.26-C2-2xF
Utilities	0.0E+00	2.8E-03	5.4E-03	1.4E-02	2.4E-03	5.0E-03	1.4E-02
Effluent	1.6E+00	1.1E+00	9.8E-01	2.0E-03	4.3E-01	3.9E-01	1.3E-03
Total	1.6E+00	1.1E+00	9.8E-01	1.6E-02	4.3E-01	3.9E-01	1.6E-02

2. *Toxicity impact assessment results for Alcalá de Henares wastewater*

2.1. Single column experiments

Table 5. Contributions to toxicity impact categories modelled with USES-LCA, from pollutants in effluents with and without treatment of Alcalá de Henares wastewater. Contributions from energy and chemicals for treatments not included here. Scores per m^3 wastewater.

Pollutants	Impact scores FAETP USES-LCA emissions to water (kg 1,4-DCB/m3 wastewater)								
	Untreated	0.05-C1	0.14-C1	0.26-C1	0.51-C1	0.77-C1	0.03-C1-2xF	0.07-C1-2xF	0.26-C1-2xF
Atenolol	4.7E-04	2.4E-04	1.7E-04	3.7E-05	1.9E-06	0.0E+00	2.8E-04	2.6E-04	5.6E-05
Bezafibrate	5.4E+00	2.4E+00	2.1E+00	1.4E+00	6.5E-01	2.8E-01	2.6E+00	2.4E+00	1.5E+00
Caffeine	7.6E-04	6.3E-04	4.7E-04	2.1E-04	8.5E-05	1.7E-05	5.1E-04	5.5E-04	2.5E-04
Carbamazepine	2.5E-05	7.0E-06	7.0E-07	0.0E+00	0.0E+00	0.0E+00	9.3E-06	7.0E-06	0.0E+00
Diclofenac	4.1E-03	0.0E+00	0.0E+00	0.0E+00	0.0E+00	0.0E+00	1.4E-03	7.9E-04	0.0E+00
Diuron	1.9E-03	0.0E+00	0.0E+00	0.0E+00	0.0E+00	0.0E+00	9.4E-04	0.0E+00	0.0E+00
Erythromycin	2.5E-01	0.0E+00	0.0E+00	0.0E+00	0.0E+00	0.0E+00	2.5E-02	0.0E+00	0.0E+00
Gemfibrozil	5.3E-02	0.0E+00	0.0E+00	0.0E+00	0.0E+00	0.0E+00	0.0E+00	0.0E+00	0.0E+00
Hydrochlorothiazide	2.2E-05	7.4E-06	4.9E-06	2.5E-07	0.0E+00	0.0E+00	9.8E-06	8.6E-06	0.0E+00
Ibuprofen	1.2E-01	6.2E-02	1.5E-02	1.5E-02	1.5E-02	0.0E+00	1.2E-01	3.1E-02	1.5E-02
N-acetyl-4-amino-antipiryne (4-AAA)	7.1E-01	2.1E-01	1.4E-02	0.0E+00	0.0E+00	0.0E+00	3.0E-01	2.5E-01	0.0E+00
Sulfamethoxazole	4.2E-02	2.8E-03	2.8E-04	0.0E+00	0.0E+00	0.0E+00	1.1E-02	5.5E-03	0.0E+00
	6.6E+00	**2.7E+00**	**2.1E+00**	**1.4E+00**	**6.7E-01**	**2.8E-01**	**3.1E+00**	**2.7E+00**	**1.5E+00**

Table 5. Cont.

Pollutants	Impact scores TETP USES-LCA emissions to soil (kg 1,4-DCB/m3 wastewater)								
	Untreated	0.05-C1	0.14-C1	0.26-C1	0.51-C1	0.77-C1	0.03-C1-2xF	0.07-C1-2xF	0.26-C1-2xF
Atenolol	1.9E-05	9.7E-06	6.7E-06	1.5E-06	7.5E-08	0.0E+00	1.1E-05	1.0E-05	2.2E-06
Bezafibrate	1.3E-03	5.8E-04	4.9E-04	3.4E-04	1.6E-04	6.7E-05	6.3E-04	5.8E-04	3.6E-04
Caffeine	2.7E-05	2.2E-05	1.6E-05	7.4E-06	3.0E-06	5.9E-07	1.8E-05	1.9E-05	8.9E-06
Carbamazepine	9.8E-08	2.7E-08	2.7E-09	0.0E+00	0.0E+00	0.0E+00	3.5E-08	2.7E-08	0.0E+00
Diclofenac	5.6E-07	0.0E+00	0.0E+00	0.0E+00	0.0E+00	0.0E+00	1.9E-07	1.1E-07	0.0E+00
Diuron	4.5E-06	0.0E+00	0.0E+00	0.0E+00	0.0E+00	0.0E+00	2.3E-06	0.0E+00	0.0E+00
Erythromycin	6.4E-04	0.0E+00	0.0E+00	0.0E+00	0.0E+00	0.0E+00	6.4E-05	0.0E+00	0.0E+00
Gemfibrozil	5.5E-06	0.0E+00	0.0E+00	0.0E+00	0.0E+00	0.0E+00	0.0E+00	0.0E+00	0.0E+00
Hydrochlorothiazide	9.2E-07	3.1E-07	2.1E-07	1.0E-08	0.0E+00	0.0E+00	4.1E-07	3.6E-07	0.0E+00
Ibuprofen	3.2E-05	1.6E-05	4.0E-06	4.0E-06	4.0E-06	0.0E+00	3.2E-05	8.0E-06	4.0E-06
N-acetyl-4-amino-antipiryne (4-AAA)	3.1E-02	9.0E-03	6.0E-04	0.0E+00	0.0E+00	0.0E+00	1.3E-02	1.1E-02	0.0E+00
Sulfamethoxazole	1.1E-03	7.0E-05	7.0E-06	0.0E+00	0.0E+00	0.0E+00	2.8E-04	1.4E-04	0.0E+00
	3.4E-02	**9.7E-03**	**1.1E-03**	**3.5E-04**	**1.6E-04**	**6.8E-05**	**1.4E-02**	**1.2E-02**	**3.7E-04**

Table 5. Cont.

Pollutants	Impact scores HTP USES-LCA emissions to soil (kg 1,4-DCB/m3 wastewater)								
	Untreated	0.05-C1	0.14-C1	0.26-C1	0.51-C1	0.77-C1	0.03-C1-2xF	0.07-C1-2xF	0.26-C1-2xF
Atenolol	1.6E-03	8.1E-04	5.6E-04	1.3E-04	6.3E-06	0.0E+00	9.4E-04	8.8E-04	1.9E-04
Bezafibrate	2.4E-04	1.1E-04	9.2E-05	6.3E-05	2.9E-05	1.3E-05	6.7E-05	1.1E-04	6.7E-05
Caffeine	6.8E-04	5.7E-04	4.2E-04	1.9E-04	7.6E-05	1.5E-05	2.3E-04	4.9E-04	2.3E-04
Carbamazepine	3.3E-04	8.9E-05	8.9E-06	0.0E+00	0.0E+00	0.0E+00	0.0E+00	8.9E-05	0.0E+00
Diclofenac	2.8E-05	0.0E+00	0.0E+00	0.0E+00	0.0E+00	0.0E+00	0.0E+00	5.4E-06	0.0E+00
Diuron	2.5E-04	0.0E+00	0.0E+00	0.0E+00	0.0E+00	0.0E+00	0.0E+00	0.0E+00	0.0E+00
Erythromycin	6.2E-05	0.0E+00	0.0E+00	0.0E+00	0.0E+00	0.0E+00	0.0E+00	0.0E+00	0.0E+00
Gemfibrozil	7.0E-02	0.0E+00	0.0E+00	0.0E+00	0.0E+00	0.0E+00	0.0E+00	0.0E+00	0.0E+00
Hydrochlorothiazide	2.3E-02	7.5E-03	5.0E-03	2.5E-04	0.0E+00	0.0E+00	0.0E+00	8.8E-03	0.0E+00
Ibuprofen	1.2E-06	6.1E-07	1.5E-07	1.5E-07	1.5E-07	0.0E+00	1.5E-07	3.0E-07	1.5E-07
N-acetyl-4-amino-antipiryne (4-AAA)	0.0E+00	0.0E+00	0.0E+00	0.0E+00	0.0E+00	0.0E+00	0.0E+00	0.0E+00	0.0E+00
Sulfamethoxazole	1.1E-04	7.5E-06	7.5E-07	0.0E+00	0.0E+00	0.0E+00	0.0E+00	1.5E-05	0.0E+00
	9.6E-02	**9.1E-03**	**6.1E-03**	**6.3E-04**	**1.1E-04**	**2.8E-05**	**1.2E-03**	**1.0E-02**	**4.8E-04**

Table 6. Contributions to toxicity impact categories modelled with EDIP'97, from pollutants in effluents with and without treatment of Alcalá de Henares wastewater. Contributions from energy and chemicals for treatments not included here. Scores per m^3 wastewater

Pollutants	Impact scores FAETP EDIP'97 emissions to water (m3 water/m3 wastewater)								
	Untreated	0.05-C1	0.14-C1	0.26-C1	0.51-C1	0.77-C1	0.03-C1-2xF	0.07-C1-2xF	0.26-C1-2xF
Atenolol	4.0E-05	2.1E-05	1.4E-05	3.2E-06	1.6E-07	0.0E+00	2.4E-05	2.2E-05	4.8E-06
Bezafibrate	8.2E-01	3.7E-01	3.1E-01	2.1E-01	9.9E-02	4.2E-02	4.0E-01	3.7E-01	2.3E-01
Caffeine	2.6E-04	2.2E-04	1.6E-04	7.3E-05	2.9E-05	5.8E-06	1.8E-04	1.9E-04	8.8E-05
Carbamazepine	4.4E-03	1.2E-03	1.2E-04	0.0E+00	0.0E+00	0.0E+00	1.6E-03	1.2E-03	0.0E+00
Diclofenac	9.1E-04	0.0E+00	0.0E+00	0.0E+00	0.0E+00	0.0E+00	3.2E-04	1.8E-04	0.0E+00
Diuron	1.0E-03	0.0E+00	0.0E+00	0.0E+00	0.0E+00	0.0E+00	5.0E-04	0.0E+00	0.0E+00
Erythromycin	2.0E-02	0.0E+00	0.0E+00	0.0E+00	0.0E+00	0.0E+00	2.0E-03	0.0E+00	0.0E+00
Gemfibrozil	7.3E-03	0.0E+00	0.0E+00	0.0E+00	0.0E+00	0.0E+00	0.0E+00	0.0E+00	0.0E+00
Hydrochlorothiazide	2.8E-06	9.5E-07	6.3E-07	3.2E-08	0.0E+00	0.0E+00	1.3E-06	1.1E-06	0.0E+00
Ibuprofen	4.8E-02	2.4E-02	5.9E-03	5.9E-03	5.9E-03	0.0E+00	4.8E-02	1.2E-02	5.9E-03
N-acetyl-4-amino-antipiryne (4-AAA)	1.2E-01	3.5E-02	2.4E-03	0.0E+00	0.0E+00	0.0E+00	5.2E-02	4.2E-02	0.0E+00
Sulfamethoxazole	7.1E-03	4.8E-04	4.8E-05	0.0E+00	0.0E+00	0.0E+00	1.9E-03	9.5E-04	0.0E+00
	1.0E+00	**4.3E-01**	**3.2E-01**	**2.2E-01**	**1.0E-01**	**4.2E-02**	**5.0E-01**	**4.2E-01**	**2.3E-01**

Table 6. Cont.

Pollutants	Impact scores TETP EDIP'97 emissions to soil (m3 soil/m3 wastewater)								
	Untreated	0.05-C1	0.14-C1	0.26-C1	0.51-C1	0.77-C1	0.03-C1-2xF	0.07-C1-2xF	0.26-C1-2xF
Atenolol	9.0E-05	4.7E-05	3.2E-05	7.2E-06	3.6E-07	0.0E+00	5.4E-05	5.0E-05	1.1E-05
Bezafibrate	1.6E+00	7.3E-01	6.2E-01	4.2E-01	2.0E-01	8.4E-02	7.8E-01	7.3E-01	4.5E-01
Caffeine	6.2E-04	5.1E-04	3.8E-04	1.7E-04	6.9E-05	1.4E-05	4.1E-04	4.5E-04	2.1E-04
Carbamazepine	5.0E-04	1.4E-04	1.4E-05	0.0E+00	0.0E+00	0.0E+00	1.8E-04	1.4E-04	0.0E+00
Diclofenac	5.2E-04	0.0E+00	0.0E+00	0.0E+00	0.0E+00	0.0E+00	1.8E-04	9.9E-05	0.0E+00
Diuron	6.8E-05	0.0E+00	0.0E+00	0.0E+00	0.0E+00	0.0E+00	3.4E-05	0.0E+00	0.0E+00
Erythromycin	5.8E-04	0.0E+00	0.0E+00	0.0E+00	0.0E+00	0.0E+00	5.8E-05	0.0E+00	0.0E+00
Gemfibrozil	7.9E-04	0.0E+00	0.0E+00	0.0E+00	0.0E+00	0.0E+00	0.0E+00	0.0E+00	0.0E+00
Hydrochlorothiazide	6.7E-06	2.2E-06	1.5E-06	7.5E-08	0.0E+00	0.0E+00	3.0E-06	2.6E-06	0.0E+00
Ibuprofen	1.8E-02	8.9E-03	2.2E-03	2.2E-03	2.2E-03	0.0E+00	1.8E-02	4.5E-03	2.2E-03
N-acetyl-4-amino-antipiryne (4-AAA)	2.9E-01	8.3E-02	5.6E-03	0.0E+00	0.0E+00	0.0E+00	1.2E-01	1.0E-01	0.0E+00
Sulfamethoxazole	1.7E-02	1.2E-03	1.2E-04	0.0E+00	0.0E+00	0.0E+00	4.6E-03	2.3E-03	0.0E+00
	2.0E+00	**8.2E-01**	**6.2E-01**	**4.2E-01**	**2.0E-01**	**8.4E-02**	**9.3E-01**	**8.4E-01**	**4.5E-01**

Table 6. Cont.

Pollutants	Impact scores HTP EDIP'97 emissions to soil (m3 soil/m3 wastewater)								
	Untreated	0.05-C1	0.14-C1	0.26-C1	0.51-C1	0.77-C1	0.03-C1-2xF	0.07-C1-2xF	0.26-C1-2xF
Atenolol	5.5E-07	2.8E-07	2.0E-07	4.4E-08	2.2E-09	0.0E+00	3.3E-07	3.1E-07	6.6E-08
Bezafibrate	5.4E-05	2.4E-05	2.0E-05	1.4E-05	6.5E-06	2.8E-06	2.6E-05	2.4E-05	1.5E-05
Caffeine	4.6E-07	3.8E-07	2.8E-07	1.3E-07	5.1E-08	1.0E-08	3.1E-07	3.3E-07	1.5E-07
Carbamazepine	1.0E-07	2.8E-08	2.8E-09	0.0E+00	0.0E+00	0.0E+00	3.7E-08	2.8E-08	0.0E+00
Diclofenac	2.9E-06	0.0E+00	0.0E+00	0.0E+00	0.0E+00	0.0E+00	9.9E-07	5.5E-07	0.0E+00
Diuron	1.3E-07	0.0E+00	0.0E+00	0.0E+00	0.0E+00	0.0E+00	6.7E-08	0.0E+00	0.0E+00
Erythromycin	8.3E-09	0.0E+00	0.0E+00	0.0E+00	0.0E+00	0.0E+00	8.3E-10	0.0E+00	0.0E+00
Gemfibrozil	6.3E-04	0.0E+00	0.0E+00	0.0E+00	0.0E+00	0.0E+00	0.0E+00	0.0E+00	0.0E+00
Hydrochlorothiazide	1.2E-05	4.0E-06	2.7E-06	1.3E-07	0.0E+00	0.0E+00	5.4E-06	4.7E-06	0.0E+00
Ibuprofen	2.0E-07	9.8E-08	2.5E-08	2.5E-08	2.5E-08	0.0E+00	2.0E-07	4.9E-08	2.5E-08
N-acetyl-4-amino-antipiryne (4-AAA)	0.0E+00	0.0E+00	0.0E+00	0.0E+00	0.0E+00	0.0E+00	0.0E+00	0.0E+00	0.0E+00
Sulfamethoxazole	1.0E-07	6.7E-09	6.7E-10	0.0E+00	0.0E+00	0.0E+00	2.7E-08	1.3E-08	0.0E+00
	7.0E-04	**2.9E-05**	**2.4E-05**	**1.4E-05**	**6.6E-06**	**2.8E-06**	**3.3E-05**	**3.0E-05**	**1.5E-05**

Table 7. Contributions to toxicity impact categories modelled with USES-LCA, from effluent (pollutants) and utilities (energy and chemicals) of Alcalá de Henares wastewater. Scores per m^3 wastewater.

FAETP USES-LCA	Untreated	0.05-C1	0.14-C1	0.26-C1	0.51-C1	0.77-C1	0.03-C1-2xF	0.07-C1-2xF	0.26-C1-2xF
Utilities	0.0E+00	8.1E-03	1.8E-02	2.6E-02	4.4E-02	5.9E-02	8.1E-03	1.8E-02	4.4E-02
Effluent	6.6E+00	2.7E+00	2.1E+00	1.4E+00	6.7E-01	2.8E-01	3.1E+00	2.7E+00	1.5E+00
Total	6.6E+00	2.7E+00	2.1E+00	1.4E+00	7.1E-01	3.4E-01	3.1E+00	2.7E+00	1.6E+00
TETP USES-LCA	Untreated	0.05-C1	0.14-C1	0.26-C1	0.51-C1	0.77-C1	0.03-C1-2xF	0.07-C1-2xF	0.26-C1-2xF
Utilities	0.0E+00	1.1E-04	2.6E-04	3.7E-04	6.2E-04	8.3E-04	1.1E-04	2.6E-04	6.2E-04
Effluent	3.4E-02	9.7E-03	1.1E-03	3.5E-04	1.6E-04	6.8E-05	1.4E-02	1.2E-02	3.7E-04
Total	3.4E-02	9.8E-03	1.4E-03	7.2E-04	7.8E-04	9.0E-04	1.4E-02	1.2E-02	9.9E-04
HTP USES-LCA	Untreated	0.05-C1	0.14-C1	0.26-C1	0.51-C1	0.77-C1	0.03-C1-2xF	0.07-C1-2xF	0.26-C1-2xF
Utilities	0.0E+00	9.8E-03	2.2E-02	3.2E-02	5.3E-02	7.1E-02	9.8E-03	2.2E-02	5.3E-02
Effluent	9.6E-02	9.1E-03	6.1E-03	6.3E-04	1.1E-04	2.8E-05	1.2E-03	1.0E-02	4.8E-04
Total	9.6E-02	1.9E-02	2.8E-02	3.3E-02	5.3E-02	7.1E-02	1.1E-02	3.2E-02	5.4E-02

Table 8. Contributions to toxicity impact categories modelled with USES-LCA, from effluent (pollutants) and utilities (energy and chemicals) of Alcalá de Henares wastewater. Scores per m^3 wastewater.

FAETP EDIP'97	Untreated	0.05-C1	0.14-C1	0.26-C1	0.51-C1	0.77-C1	0.03-C1-2xF	0.07-C1-2xF	0.26-C1-2xF
Utilities	0.0E+00	6.4E+01	1.2E+02	1.9E+02	3.2E+02	4.7E+02	6.5E+01	1.2E+02	3.2E+02
Effluent	1.0E+03	4.3E+02	3.2E+02	2.2E+02	1.0E+02	4.2E+01	5.0E+02	4.2E+02	2.3E+02
Total	1.0E+03	4.9E+02	4.4E+02	4.1E+02	4.2E+02	5.1E+02	5.7E+02	5.4E+02	5.5E+02
TETP EDIP'97	Untreated	0.05-C1	0.14-C1	0.26-C1	0.51-C1	0.77-C1	0.03-C1-2xF	0.07-C1-2xF	0.26-C1-2xF
Utilities	0.0E+00	5.0E-01	9.4E-01	1.5E+00	2.5E+00	3.6E+00	5.0E-01	9.3E-01	2.5E+00
Effluent	2.0E+03	8.2E+02	6.2E+02	4.2E+02	2.0E+02	8.4E+01	9.3E+02	8.4E+02	4.5E+02
Total	2.0E+03	8.2E+02	6.3E+02	4.2E+02	2.0E+02	8.8E+01	9.3E+02	8.4E+02	4.5E+02
HTP EDIP'97	Untreated	0.05-C1	0.14-C1	0.26-C1	0.51-C1	0.77-C1	0.03-C1-2xF	0.07-C1-2xF	0.26-C1-2xF
Utilities	0.0E+00	2.8E-03	5.3E-03	8.4E-03	1.4E-02	2.0E-02	2.8E-03	5.3E-03	1.4E-02
Effluent	7.0E-01	2.9E-02	2.4E-02	1.4E-02	6.6E-03	2.8E-03	3.3E-02	3.0E-02	1.5E-02
Total	7.0E-01	3.2E-02	2.9E-02	2.3E-02	2.0E-02	2.3E-02	3.6E-02	3.5E-02	2.9E-02

2.2. Double column experiments

Table 9. Contributions to toxicity impact categories modelled with USES-LCA, from pollutants in effluents with and without treatment of Alcalá de Henares wastewater. Contributions from energy and chemicals for treatments not included here. Scores per m^3 wastewater.

Pollutants	Impact scores FAETP USES-LCA emissions to water (kg 1,4-DCB/m3 wastewater)								
	Untreated	0.05-C2	0.14-C2	0.26-C2	0.51-C2	0.77-C2	0.03-C2-2xF	0.07-C2-2xF	0.26-C2-2xF
Atenolol	4.7E-04	2.4E-04	1.7E-04	3.7E-05	1.9E-06	0.0E+00	2.8E-04	2.6E-04	5.6E-05
Bezafibrate	5.4E+00	2.4E+00	2.1E+00	1.4E+00	6.5E-01	2.8E-01	2.6E+00	2.4E+00	1.5E+00
Caffeine	7.6E-04	6.3E-04	4.7E-04	2.1E-04	8.5E-05	1.7E-05	5.1E-04	5.5E-04	2.5E-04
Carbamazepine	2.5E-05	7.0E-06	7.0E-07	0.0E+00	0.0E+00	0.0E+00	9.3E-06	7.0E-06	0.0E+00
Diclofenac	4.1E-03	0.0E+00	0.0E+00	0.0E+00	0.0E+00	0.0E+00	1.4E-03	7.9E-04	0.0E+00
Diuron	1.9E-03	0.0E+00	0.0E+00	0.0E+00	0.0E+00	0.0E+00	9.4E-04	0.0E+00	0.0E+00
Erythromycin	2.5E-01	0.0E+00	0.0E+00	0.0E+00	0.0E+00	0.0E+00	2.5E-02	0.0E+00	0.0E+00
Gemfibrozil	5.3E-02	0.0E+00	0.0E+00	0.0E+00	0.0E+00	0.0E+00	0.0E+00	0.0E+00	0.0E+00
Hydrochlorothiazide	2.2E-05	7.4E-06	4.9E-06	2.5E-07	0.0E+00	0.0E+00	9.8E-06	8.6E-06	0.0E+00
Ibuprofen	1.2E-01	6.2E-02	1.5E-02	1.5E-02	1.5E-02	0.0E+00	1.2E-01	3.1E-02	1.5E-02
N-acetyl-4-amino-antipiryne (4-AAA)	7.1E-01	2.1E-01	1.4E-02	0.0E+00	0.0E+00	0.0E+00	3.0E-01	2.5E-01	0.0E+00
Sulfamethoxazole	4.2E-02	2.8E-03	2.8E-04	0.0E+00	0.0E+00	0.0E+00	1.1E-02	5.5E-03	0.0E+00
	6.6E+00	**2.7E+00**	**2.1E+00**	**1.4E+00**	**6.7E-01**	**2.8E-01**	**3.1E+00**	**2.7E+00**	**1.5E+00**

Table 9. Cont.

Pollutants	Impact scores TETP USES-LCA emissions to soil (kg 1,4-DCB/m3 wastewater)								
	Untreated	0.05-C2	0.14-C2	0.26-C2	0.51-C2	0.77-C2	0.03-C2-2xF	0.07-C2-2xF	0.26-C2-2xF
Atenolol	1.9E-05	9.7E-06	6.7E-06	1.5E-06	7.5E-08	0.0E+00	1.1E-05	1.0E-05	2.2E-06
Bezafibrate	1.3E-03	5.8E-04	4.9E-04	3.4E-04	1.6E-04	6.7E-05	6.3E-04	5.8E-04	3.6E-04
Caffeine	2.7E-05	2.2E-05	1.6E-05	7.4E-06	3.0E-06	5.9E-07	1.8E-05	1.9E-05	8.9E-06
Carbamazepine	9.8E-08	2.7E-08	2.7E-09	0.0E+00	0.0E+00	0.0E+00	3.5E-08	2.7E-08	0.0E+00
Diclofenac	5.6E-07	0.0E+00	0.0E+00	0.0E+00	0.0E+00	0.0E+00	1.9E-07	1.1E-07	0.0E+00
Diuron	4.5E-06	0.0E+00	0.0E+00	0.0E+00	0.0E+00	0.0E+00	2.3E-06	0.0E+00	0.0E+00
Erythromycin	6.4E-04	0.0E+00	0.0E+00	0.0E+00	0.0E+00	0.0E+00	6.4E-05	0.0E+00	0.0E+00
Gemfibrozil	5.5E-06	0.0E+00	0.0E+00	0.0E+00	0.0E+00	0.0E+00	0.0E+00	0.0E+00	0.0E+00
Hydrochlorothiazide	9.2E-07	3.1E-07	2.1E-07	1.0E-08	0.0E+00	0.0E+00	4.1E-07	3.6E-07	0.0E+00
Ibuprofen	3.2E-05	1.6E-05	4.0E-06	4.0E-06	4.0E-06	0.0E+00	3.2E-05	8.0E-06	4.0E-06
N-acetyl-4-amino-antipiryne (4-AAA)	3.1E-02	9.0E-03	6.0E-04	0.0E+00	0.0E+00	0.0E+00	1.3E-02	1.1E-02	0.0E+00
Sulfamethoxazole	1.1E-03	7.0E-05	7.0E-06	0.0E+00	0.0E+00	0.0E+00	2.8E-04	1.4E-04	0.0E+00
	3.4E-02	**9.7E-03**	**1.1E-03**	**3.5E-04**	**1.6E-04**	**6.8E-05**	**1.4E-02**	**1.2E-02**	**3.7E-04**

Table 9. Cont.

Pollutants	Impact scores HTP USES-LCA emissions to soil (kg 1,4-DCB/m3 wastewater)								
	Untreated	0.05-C2	0.14-C2	0.26-C2	0.51-C2	0.77-C2	0.03-C2-2xF	0.07-C2-2xF	0.26-C2-2xF
Atenolol	1.6E-03	8.1E-04	5.6E-04	1.3E-04	6.3E-06	0.0E+00	9.4E-04	8.8E-04	1.9E-04
Bezafibrate	2.4E-04	1.1E-04	9.2E-05	6.3E-05	2.9E-05	1.3E-05	1.2E-04	1.1E-04	6.7E-05
Caffeine	6.8E-04	5.7E-04	4.2E-04	1.9E-04	7.6E-05	1.5E-05	4.6E-04	4.9E-04	2.3E-04
Carbamazepine	3.3E-04	8.9E-05	8.9E-06	0.0E+00	0.0E+00	0.0E+00	1.2E-04	8.9E-05	0.0E+00
Diclofenac	2.8E-05	0.0E+00	0.0E+00	0.0E+00	0.0E+00	0.0E+00	9.7E-06	5.4E-06	0.0E+00
Diuron	2.5E-04	0.0E+00	0.0E+00	0.0E+00	0.0E+00	0.0E+00	1.3E-04	0.0E+00	0.0E+00
Erythromycin	6.2E-05	0.0E+00	0.0E+00	0.0E+00	0.0E+00	0.0E+00	6.2E-06	0.0E+00	0.0E+00
Gemfibrozil	7.0E-02	0.0E+00	0.0E+00	0.0E+00	0.0E+00	0.0E+00	0.0E+00	0.0E+00	0.0E+00
Hydrochlorothiazide	2.3E-02	7.5E-03	5.0E-03	2.5E-04	0.0E+00	0.0E+00	1.0E-02	8.8E-03	0.0E+00
Ibuprofen	1.2E-06	6.1E-07	1.5E-07	1.5E-07	1.5E-07	0.0E+00	1.2E-06	3.0E-07	1.5E-07
N-acetyl-4-amino-antipiryne (4-AAA)	0.0E+00	0.0E+00	0.0E+00	0.0E+00	0.0E+00	0.0E+00	0.0E+00	0.0E+00	0.0E+00
Sulfamethoxazole	1.1E-04	7.5E-06	7.5E-07	0.0E+00	0.0E+00	0.0E+00	3.0E-05	1.5E-05	0.0E+00
	9.6E-02	**9.1E-03**	**6.1E-03**	**6.3E-04**	**1.1E-04**	**2.8E-05**	**1.2E-02**	**1.0E-02**	**4.8E-04**

Table 10. Contributions to toxicity impact categories modelled with EDIP'97, from pollutants in effluents with and without treatment of Alcalá de Henares wastewater. Contributions from energy and chemicals for treatments not included here. Scores per m^3 wastewater.

Pollutants	Impact scores FAETP EDIP'97 emissions to water (m3 water/m3 wastewater)								
	Untreated	0.05-C2	0.14-C2	0.26-C2	0.51-C2	0.77-C2	0.03-C2-2xF	0.07-C2-2xF	0.26-C2-2xF
Atenolol	4.0E-02	2.1E-02	1.4E-02	3.2E-03	1.6E-04	0.0E+00	2.4E-02	2.2E-02	4.8E-03
Bezafibrate	8.2E+02	3.7E+02	3.1E+02	2.1E+02	9.9E+01	4.2E+01	4.0E+02	3.7E+02	2.3E+02
Caffeine	2.6E-01	2.2E-01	1.6E-01	7.3E-02	2.9E-02	5.8E-03	1.8E-01	1.9E-01	8.8E-02
Carbamazepine	4.4E+00	1.2E+00	1.2E-01	0.0E+00	0.0E+00	0.0E+00	1.6E+00	1.2E+00	0.0E+00
Diclofenac	9.1E-01	0.0E+00	0.0E+00	0.0E+00	0.0E+00	0.0E+00	3.2E-01	1.8E-01	0.0E+00
Diuron	1.0E+00	0.0E+00	0.0E+00	0.0E+00	0.0E+00	0.0E+00	5.0E-01	0.0E+00	0.0E+00
Erythromycin	2.0E+01	0.0E+00	0.0E+00	0.0E+00	0.0E+00	0.0E+00	2.0E+00	0.0E+00	0.0E+00
Gemfibrozil	7.3E+00	0.0E+00	0.0E+00	0.0E+00	0.0E+00	0.0E+00	0.0E+00	0.0E+00	0.0E+00
Hydrochlorothiazide	2.8E-03	9.5E-04	6.3E-04	3.2E-05	0.0E+00	0.0E+00	1.3E-03	1.1E-03	0.0E+00
Ibuprofen	4.8E+01	2.4E+01	5.9E+00	5.9E+00	5.9E+00	0.0E+00	4.8E+01	1.2E+01	5.9E+00
N-acetyl-4-amino-antipiryne (4-AAA)	1.2E+02	3.5E+01	2.4E+00	0.0E+00	0.0E+00	0.0E+00	5.2E+01	4.2E+01	0.0E+00
Sulfamethoxazole	7.1E+00	4.8E-01	4.8E-02	0.0E+00	0.0E+00	0.0E+00	1.9E+00	9.5E-01	0.0E+00
	1.0E+03	**4.3E+02**	**3.2E+02**	**2.2E+02**	**1.0E+02**	**4.2E+01**	**5.0E+02**	**4.2E+02**	**2.3E+02**

Table 10. Cont.

Pollutants	Impact scores TETP EDIP'97 emissions to soil (m3 soil/m3 wastewater)								
	Untreated	0.05-C2	0.14-C2	0.26-C2	0.51-C2	0.77-C2	0.03-C2-2xF	0.07-C2-2xF	0.26-C2-2xF
Atenolol	9.0E-02	4.7E-02	3.2E-02	7.2E-03	3.6E-04	0.0E+00	5.4E-02	5.0E-02	1.1E-02
Bezafibrate	1.6E+03	7.3E+02	6.2E+02	4.2E+02	2.0E+02	8.4E+01	7.8E+02	7.3E+02	4.5E+02
Caffeine	6.2E-01	5.1E-01	3.8E-01	1.7E-01	6.9E-02	1.4E-02	4.1E-01	4.5E-01	2.1E-01
Carbamazepine	5.0E-01	1.4E-01	1.4E-02	0.0E+00	0.0E+00	0.0E+00	1.8E-01	1.4E-01	0.0E+00
Diclofenac	5.2E-01	0.0E+00	0.0E+00	0.0E+00	0.0E+00	0.0E+00	1.8E-01	9.9E-02	0.0E+00
Diuron	6.8E-02	0.0E+00	0.0E+00	0.0E+00	0.0E+00	0.0E+00	3.4E-02	0.0E+00	0.0E+00
Erythromycin	5.8E-01	0.0E+00	0.0E+00	0.0E+00	0.0E+00	0.0E+00	5.8E-02	0.0E+00	0.0E+00
Gemfibrozil	7.9E-01	0.0E+00	0.0E+00	0.0E+00	0.0E+00	0.0E+00	0.0E+00	0.0E+00	0.0E+00
Hydrochlorothiazide	6.7E-03	2.2E-03	1.5E-03	7.5E-05	0.0E+00	0.0E+00	3.0E-03	2.6E-03	0.0E+00
Ibuprofen	1.8E+01	8.9E+00	2.2E+00	2.2E+00	2.2E+00	0.0E+00	1.8E+01	4.5E+00	2.2E+00
N-acetyl-4-amino-antipiryne (4-AAA)	2.9E+02	8.3E+01	5.6E+00	0.0E+00	0.0E+00	0.0E+00	1.2E+02	1.0E+02	0.0E+00
Sulfamethoxazole	1.7E+01	1.2E+00	1.2E-01	0.0E+00	0.0E+00	0.0E+00	4.6E+00	2.3E+00	0.0E+00
	2.0E+03	**8.2E+02**	**6.2E+02**	**4.2E+02**	**2.0E+02**	**8.4E+01**	**9.3E+02**	**8.4E+02**	**4.5E+02**

Table 10. Cont.

Pollutants	Impact scores HTP EDIP'97 emissions to soil (m3 soil/m3 wastewater)								
	Untreated	0.05-C2	0.14-C2	0.26-C2	0.51-C2	0.77-C2	0.03-C2-2xF	0.07-C2-2xF	0.26-C2-2xF
Atenolol	5.5E-04	2.8E-04	2.0E-04	4.4E-05	2.2E-06	0.0E+00	3.3E-04	3.1E-04	6.6E-05
Bezafibrate	5.4E-02	2.4E-02	2.0E-02	1.4E-02	6.5E-03	2.8E-03	2.6E-02	2.4E-02	1.5E-02
Caffeine	4.6E-04	3.8E-04	2.8E-04	1.3E-04	5.1E-05	1.0E-05	3.1E-04	3.3E-04	1.5E-04
Carbamazepine	1.0E-04	2.8E-05	2.8E-06	0.0E+00	0.0E+00	0.0E+00	3.7E-05	2.8E-05	0.0E+00
Diclofenac	2.9E-03	0.0E+00	0.0E+00	0.0E+00	0.0E+00	0.0E+00	9.9E-04	5.5E-04	0.0E+00
Diuron	1.3E-04	0.0E+00	0.0E+00	0.0E+00	0.0E+00	0.0E+00	6.7E-05	0.0E+00	0.0E+00
Erythromycin	8.3E-06	0.0E+00	0.0E+00	0.0E+00	0.0E+00	0.0E+00	8.3E-07	0.0E+00	0.0E+00
Gemfibrozil	6.3E-01	0.0E+00	0.0E+00	0.0E+00	0.0E+00	0.0E+00	0.0E+00	0.0E+00	0.0E+00
Hydrochlorothiazide	1.2E-02	4.0E-03	2.7E-03	1.3E-04	0.0E+00	0.0E+00	5.4E-03	4.7E-03	0.0E+00
Ibuprofen	2.0E-04	9.8E-05	2.5E-05	2.5E-05	2.5E-05	0.0E+00	2.0E-04	4.9E-05	2.5E-05
N-acetyl-4-amino-antipiryne (4-AAA)	0.0E+00	0.0E+00	0.0E+00	0.0E+00	0.0E+00	0.0E+00	0.0E+00	0.0E+00	0.0E+00
Sulfamethoxazole	1.0E-04	6.7E-06	6.7E-07	0.0E+00	0.0E+00	0.0E+00	2.7E-05	1.3E-05	0.0E+00
	7.0E-01	**2.9E-02**	**2.4E-02**	**1.4E-02**	**6.6E-03**	**2.8E-03**	**3.3E-02**	**3.0E-02**	**1.5E-02**

Table 11. Contributions to toxicity impact categories modelled with USES-LCA, from effluent (pollutants) and utilities (energy and chemicals) of Alcalá de Henares wastewater. Scores per m^3 wastewater.

FAETP USES-LCA	Untreated	0.05-C2	0.14-C2	0.26-C2	0.51-C2	0.77-C2	0.03-C2-2xF	0.07-C2-2xF	0.26-C2-2xF
Utilities	0.0E+00	9.1E-03	1.7E-02	2.7E-02	4.5E-02	6.6E-02	9.2E-03	1.7E-02	4.5E-02
Effluent	6.6E+00	2.7E+00	2.1E+00	1.4E+00	6.7E-01	2.8E-01	3.1E+00	2.7E+00	1.5E+00
Total	6.6E+00	2.7E+00	2.1E+00	1.4E+00	7.2E-01	3.5E-01	3.1E+00	2.7E+00	1.6E+00
TETP USES-LCA	Untreated	0.05-C2	0.14-C2	0.26-C2	0.51-C2	0.77-C2	0.03-C2-2xF	0.07-C2-2xF	0.26-C2-2xF
Utilities	0.0E+00	1.3E-04	2.4E-04	3.9E-04	6.3E-04	9.3E-04	1.3E-04	2.4E-04	6.3E-04
Effluent	3.4E-02	9.7E-03	1.1E-03	3.5E-04	1.6E-04	6.8E-05	1.4E-02	1.2E-02	3.7E-04
Total	3.4E-02	9.9E-03	1.4E-03	7.4E-04	8.0E-04	1.0E-03	1.4E-02	1.2E-02	1.0E-03
HTP USES-LCA	Untreated	0.05-C2	0.14-C2	0.26-C2	0.51-C2	0.77-C2	0.03-C2-2xF	0.07-C2-2xF	0.26-C2-2xF
Utilities	0.0E+00	1.1E-02	2.1E-02	3.3E-02	5.4E-02	8.0E-02	1.1E-02	2.1E-02	5.4E-02
Effluent	9.6E-02	9.1E-03	6.1E-03	6.3E-04	1.1E-04	2.8E-05	1.2E-02	1.0E-02	4.8E-04
Total	9.6E-02	2.0E-02	2.7E-02	3.4E-02	5.5E-02	8.0E-02	2.3E-02	3.1E-02	5.5E-02

Table 12. Contributions to toxicity impact categories modelled with EDIP'97, from effluent (pollutants) and utilities (energy and chemicals) of Alcalá de Henares wastewater. Scores per m^3 wastewater.

FAETP EDIP'97	Untreated	0.05-C2	0.14-C2	0.26-C2	0.51-C2	0.77-C2	0.03-C2-2xF	0.07-C2-2xF	0.26-C2-2xF
Utilities	0.0E+00	6.4E+01	1.2E+02	1.9E+02	3.2E+02	4.7E+02	6.5E+01	1.2E+02	3.2E+02
Effluent	1.0E+03	4.3E+02	3.2E+02	2.2E+02	1.0E+02	4.2E+01	5.0E+02	4.2E+02	2.3E+02
Total	1.0E+03	4.9E+02	4.4E+02	4.1E+02	4.2E+02	5.1E+02	5.7E+02	5.4E+02	5.5E+02
TETP EDIP'97	Untreated	0.05-C2	0.14-C2	0.26-C2	0.51-C2	0.77-C2	0.03-C2-2xF	0.07-C2-2xF	0.26-C2-2xF
Utilities	0.0E+00	5.0E-01	9.4E-01	1.5E+00	2.5E+00	3.6E+00	5.0E-01	9.3E-01	2.5E+00
Effluent	2.0E+03	8.2E+02	6.2E+02	4.2E+02	2.0E+02	8.4E+01	9.3E+02	8.4E+02	4.5E+02
Total	2.0E+03	8.2E+02	6.3E+02	4.2E+02	2.0E+02	8.8E+01	9.3E+02	8.4E+02	4.5E+02
HTP EDIP'97	Untreated	0.05-C2	0.14-C2	0.26-C2	0.51-C2	0.77-C2	0.03-C2-2xF	0.07-C2-2xF	0.26-C2-2xF
Utilities	0.0E+00	2.8E-03	5.3E-03	8.4E-03	1.4E-02	2.0E-02	2.8E-03	5.3E-03	1.4E-02
Effluent	7.0E-01	2.9E-02	2.4E-02	1.4E-02	6.6E-03	2.8E-03	3.3E-02	3.0E-02	1.5E-02
Total	7.0E-01	3.2E-02	2.9E-02	2.3E-02	2.0E-02	2.3E-02	3.6E-02	3.5E-02	2.9E-02