



Research article

Life cycle environmental impacts of advanced wastewater treatment techniques for removal of pharmaceuticals and personal care products (PPCPs)

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ABSTRACT

Pharmaceutical and personal care products (PPCPs) are of increasing interest because of their ecotoxicological properties and environmental impacts. Wastewater treatment plants (WWTPs) are the main pathway for their release into freshwaters due to the inefficiency of conventional WWTPs in removing many of these contaminants from effluents. Therefore, different advanced effluent treatment techniques have been proposed for their treatment. However, it is not known at present how effective these treatment methods are and whether on a life cycle basis they cause other environmental impacts which may outweigh the benefits of the treatment. In an effort to provide an insight into this question, this paper considers life cycle environmental impacts of the following advanced treatment techniques aimed at reducing freshwater ecotoxicity potential of PPCPs: granular activated carbon (GAC), nanofiltration (NF), solar photo-Fenton (SPF) and ozonation. The results suggest that on average NF has the lowest impacts for 13 out of 18 categories considered. GAC is the best alternative for five impacts, including metals and water depletion, but it has the highest marine eutrophication. SPF and ozonation are the least sustainable for eight impacts, including ecotoxicity and climate change. GAC and NF are also more efficient in treating heavy metals while avoiding generation of harmful by-products during the treatment, thus being more suitable for potable reuse of wastewater. However, releasing the effluent without advanced treatment to agricultural land achieves a much higher reduction of freshwater ecotoxicity than treating it by any of the advanced treatments and releasing to the environment. Therefore, the use of advanced effluent treatment for agricultural purposes is not recommended.

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1. Introduction

Ageing population, increasing urbanisation, growing per-capita income and other factors have led to a significant increase in the consumption of pharmaceutical and personal care products (PPCPs) and this trend is expected to continue in the future (Hill and Chu, 2009; Lyons, 2014; ter Laak et al., 2010; WHO, 2004). Over 3000 chemicals are used in the production of PPCPs (Daughton, 2004) and they eventually reach the environment. One of the main pathways for this are wastewater treatment plants (WWTPs) (Celle-jeanton et al., 2014; Heberer, 2002; Onesios et al., 2009). This is due to the inefficiency of conventional plants in removing many

PPCPs as they are designed to treat biodegradable, non-polar and large molecules, as opposed to small molecules and polar tendency of PPCP substances (Daughton, 2001; Ratola et al., 2012; Richardson et al., 2005; Suárez et al., 2008). Additionally, several metabolites and transformation products are also expected to form in WWTP effluents alongside parent compounds as a result of various chemical reactions during the treatment (Farré et al., 2008; Quintana et al., 2005; Schuman, 2008).

However, despite their widespread presence in the environment, often at notable concentrations (Carmona et al., 2014; Hughes et al., 2013; Kolpin et al., 2002), relatively little is known about their ecotoxicological effects (Boxall, 2004; Carlsson et al., 2006; Fick et al., 2009; Jones-Lepp and Stevens, 2007; Khetan and Collins, 2007). To address the emerging concerns related to the impacts of these contaminants on the environment (Blasco and Delvalls, 2008; Clevers, 2004; Fedorenkova et al., 2010; Fent et al.,

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2006), legislation that requires their removal from WWTP effluents is currently being developed in Europe (Eggen et al., 2014; European Commission, 2015). Thus, it is necessary to consider possible advanced treatment options that could help reduce their release into the environment (Bui et al., 2016). A further motivation for considering such options is that water will become increasingly scarce in many world regions and wastewater reuse is being advocated as an option for increasing its availability (Miller, 2006; Rodríguez et al., 2009; Urkiaga et al., 2006). Thereby, regulations towards emerging contaminants are under discussion to ensure safer wastewater reuse (Dalahmeh and Baresel, 2014; EPA, 2012; Hanjra et al., 2012). To this end, some of the advanced effluent techniques are capable of not only removing PPCPs but could also enable reuse of water, either for irrigation or drinking (Bixio et al., 2008; CDPH, 2009; Tchobanoglous et al., 2011).

At present, however, it is not known how environmentally effective different advanced treatment methods are in reducing the ecotoxicity impacts of PPCP compounds and if, by doing so, they increase the ecotoxicity potential elsewhere in the life cycle or cause other environmental impacts (Kärman, 2001; Muga and Mihelcic, 2008; Pasqualino et al., 2011; Tangsubkul et al., 2005). In an attempt to inform this debate, this paper considers life cycle environmental impacts of the following advanced treatment methods: granular activated carbon (GAC), nanofiltration (NF), solar photo-Fenton (SPF) and ozonation. These methods have been chosen for consideration because they are either already well established (the first two) or are most promising techniques (the last two) for removal of PPCPs.

As far as the authors are aware, this is the first study of its kind, providing a comprehensive quantitative comparison of these methods for a range of operating parameters typically found in WWTPs. Furthermore, no other studies considered the ecotoxicity potential of the PPCP compounds when released to freshwaters or used for agricultural purposes without advanced treatment in comparison with the ecotoxicity potential when treated in advanced treatment systems.

2. Methods

Life cycle assessment (LCA) has been used as a tool to estimate life cycle impacts of the chosen treatment methods following the ISO 14040 methodology (ISO, 2006). The next sections detail the goal and scope together with the assumptions and data used in the study.

2.1. Goal and scope of the study

The goal of the study was to estimate and compare life cycle environmental impacts of the four advanced techniques – GAC, NF, SPF and ozonation – for the treatment of PPCPs. A further goal was to estimate the ecotoxicity of PPCPs in the effluent after advanced treatment and compare it to the equivalent impact of the effluent from conventional WWTPs without advanced treatment. The following nine PPCPs were chosen for consideration, spanning five product categories:

- analgesics: diclofenac and ibuprofen;
- antibiotics: erythromycin, trimethoprim and sulfamethoxazole;
- psychiatric drugs: carbamazepine;
- hormones: estrone and 17 β -estradiol; and
- antiseptics: triclosan.

These substances were selected due to:

- their potential to cause undesired ecotoxicological effects;
- higher concentration and frequency of detection in effluents of conventional WWTPs; and
- availability of ecotoxicological data.

The system and the system boundaries are outlined in Fig. 1. As indicated in the figure, the scope of the study was from 'cradle to grave', encompassing construction, operation and decommissioning of the treatment plant. The advanced effluent treatment plant was assumed to be coupled with a conventional WWTP, which includes a membrane bioreactor (MBR) as part of the secondary activated sludge treatment. The MBR is needed to enable a more efficient operation of the advanced treatment plant because it reduces total suspended solids and biochemical oxygen demand more efficiently than conventional biological treatment (Alturki et al., 2010; Cases et al., 2011; Laera et al., 2012; Snyder et al., 2007). The assumed capacity of the MBR plant was 64,000 m³/d, corresponding to the average capacity of WWTPs in the UK (DEFRA, 2012). Furthermore, it was assumed that the advanced treatment methods are capable of treating wastewater to near potable water standards since they are controlled for pH, pathogens, hardness and heavy metals.

The functional unit was defined as the "treatment of 1000 m³ of effluent from conventional wastewater treatment". The treatment facility was assumed to be located in the UK. The lifetime of the plants was assumed at 60 years (Pasqualino et al., 2009), with periodic replacements of equipment, as detailed further below.

2.2. Inventory data

The inventory data were sourced from the literature and own estimates as detailed in the next sections. Industry data were not available as the removal of PPCP compounds is still not carried out by the water industry due to a lack of legislation. The life cycle data were taken from Ecoinvent 2.2 (Frischknecht et al., 2004) and the UK electricity grid updated to the year 2016 (BEIS, 2016).

The following sections give a brief description of the advanced treatment methods considered, followed by an overview of the estimation of the operating parameters.

2.2.1. Overview of advanced treatment techniques

2.2.1.1. Granular activated carbon. GAC treatment removes PPCP compounds by physical adsorption onto the GAC bed and to a lesser

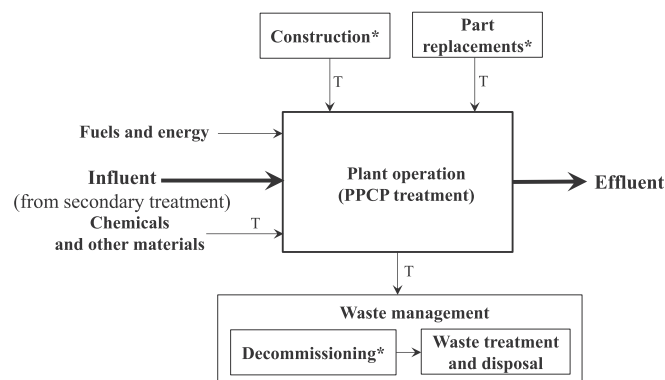


Fig. 1. System boundaries and life cycle stages of the advanced effluent treatments techniques considered in the study (*Excluded for ozonation due to a lack of data. T: transport).

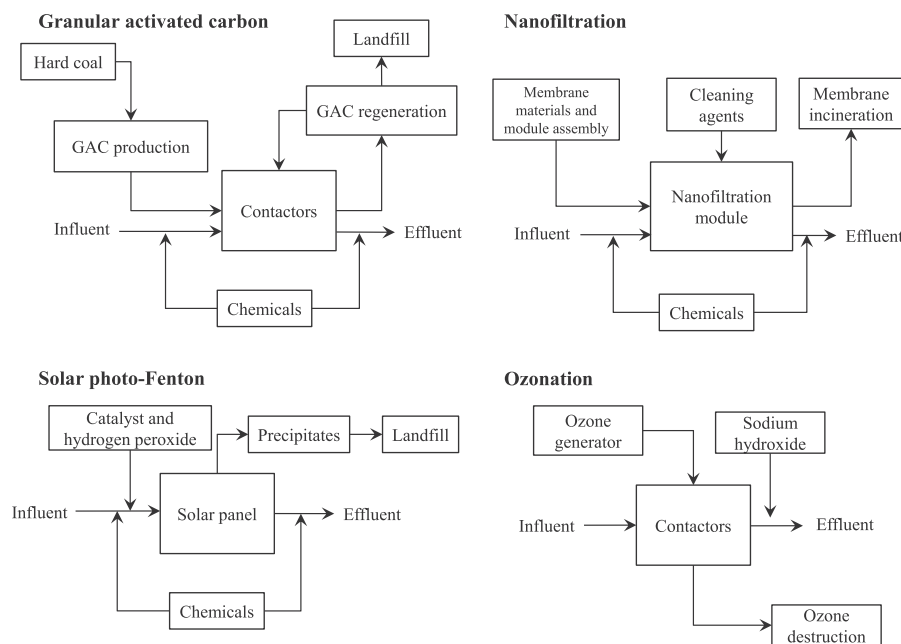


Fig. 2. An overview of the advanced PPCP treatment methods considered in the study.

extent through biodegradation, thus avoiding generation of harmful reaction by-products. Moreover, a high removal of metals is expected (Goel et al., 2005; Fu and Wang, 2011). After a certain time in use, the bed needs to be regenerated or replaced (Fig. 2). Hence, the key influencing parameters are the amount of fresh GAC required for the treatment and the number of regeneration cycles before the bed needs to be replaced. These were estimated according to the method detailed in section 2.2.2.1.

2.2.1.2. Nanofiltration. NF uses membranes to remove PPCP compounds from the effluent (Fig. 2). First, the wastewater is passed through pre-filters under high pressure to remove particles and then through the membranes with the pore sizes of 0.1–1.0 nm, separating the influx into the permeate (treated effluent) and concentrate (contaminants), assumed to be recycled back to the treatment. The primary removal of contaminants is through physical sieving, followed by adsorption and electrostatic repulsion, which can achieve a high removal efficiency of heavy metals and avoid the formation of by-products (Bellona et al., 2004; Bolong et al., 2009; Fu and Wang, 2011). NF units are designed in a modular fashion, with the total number of modules varying according to the influent flow and the desired quality of effluent. One important factor in NF treatments is membrane fouling, causing a pressure drop and requiring more electricity to maintain the flux. When the pressure drop becomes too large, the units need to be cleaned using chemical agents (Al-Amoudi and Lovitt, 2007; Simon et al., 2013). Therefore, the key operating parameters that need to be considered for NF are electricity consumption and the usage of cleaning agents. The former includes electricity for pre-filtration and NF, high-pressure pumping, water heating during winter months and the lighting of the building. The type of chemical agents used depends on the influent composition, filtration pressure and membrane properties (Bruggen et al., 2008; Yoon et al., 2006). The data for NF were taken from real facilities in Canada and France (Bonton et al., 2012; Cyna et al., 2002), as detailed in section 2.2.4.

2.2.1.3. Solar photo-Fenton. The solar-photo Fenton (SPF) technique is an advanced oxidation process known for simple operation and high efficiency in treating most organic contaminants. The process consists of adding a catalyst and hydrogen peroxide to the influent which is then passed through reactors irradiated by solar light to generate OH radicals and oxidise PPCPs (Fig. 2). To increase the treatment efficiency, the influent should be kept at low pH, followed by its neutralisation before the discharge into the environment (Muñoz et al., 2007). Hence, the main parameters that need to be considered in this treatment are the amounts of the catalyst and hydrogen peroxide. These are still subject of ongoing research as SPF is still under development, but some pilot-plant data are available (Klamerth, 2011; Lofrano, 2012; Ribeiro et al., 2015) which were considered here. These and other inventory data for SPF can be found in section 2.2.4.

2.2.1.4. Ozonation. The ozonation treatment (disinfection), works through direct and indirect reactions of PPCPs with OH radicals generated by ozone decomposition in the contactors (Fig. 2). This method can achieve a good removal of metals but can also generate harmful by-products (Margot et al., 2013; Tripathi and Tripathi, 2011; Westerhoff et al., 2005). The overall treatment efficiency is directly dependent on the influent pH, contact time and organic matter content. After the treatment, the effluent is balanced by the addition of sodium hydroxide. The ozone can be generated either from liquid oxygen or air, with the latter mainly used at large-scale facilities; both methods have significant electricity requirements (Kim and Tanaka, 2011; Wang et al., 2005). The main parameters that need to be considered in the design of ozonation units are the amount of ozone required for efficient treatment and electricity consumption. Their estimation is discussed in section 2.2.2.2.

2.2.2. Estimation of operating parameters for GAC and ozonation

As mentioned above, data for the operating parameters for NF and SPF were taken from the literature and are discussed in section 2.2.4. However, for GAC and ozonation data for the key operating

Table 1Operating parameters for GAC (eqns. (1)–(4)) and ozonation (eqns. (5) and (6)) per 1000 m³ of wastewater.

	EBCT (min)	Q _{inf} (m ³ /h)	V _{GAC} (m ³)	m _{GAC} ^a (kg)	t _{GAC} ^b (d)	N _{BR} ^c	n _r	Fresh GAC ^d (kg)	Regenerated GAC (kg)
GAC	20	2667	889	501,396	330	6	6	6,818,986	33,092,136
	30		1334	752,094	220	9	9	14,966,671	74,457,306
	40		1778	1,002,792	110	19	9	40,011,401	199,555,608
Ozonation	T (kg/m ³)	TE (%)	D _{ozone} (kg/m ³)	V _{inf} (m ³)	E _{ozone} (kWh/kg _{ozone})	E _{ozonation} (kWh)			
	0.004	75	0.005	1000	16.5	88			
	0.023	50	0.046			759			
	0.042	25	0.168			2772			

^a GAC density: 564 kg/m³.^b Values from Fig. 3.^c n_{max} = 10; T_{treatment}: 21,900 days (over the 60 year lifespan).^d m_{loss} = 10% (Bayer et al., 2005).

parameters were not available and had to be estimated as detailed below.

2.2.2.1. Granular activated carbon. The key operating parameters for GAC – the amount of fresh GAC and the number of regeneration cycles before the bed has to be replaced – were estimated based on two criteria commonly considered in the design of GAC: empty-bed contact time (EBCT) and the bed service time (Bayer et al., 2005; Creek and Davidson, 2000; Reed et al., 1996). The initial amount of fresh granular activated carbon was estimated for different EBCTs, as follows:

$$V_{GAC} = EBCT \times Q_{inf} \quad (m^3) \quad (1)$$

where:

V_{GAC} volume of granular activated carbon in the bed (m³)

EBCT empty-bed contact time (min)

Q_{inf} influent flow to be treated (m³/min).

The maximum number of bed regenerations was fixed at 10 to ensure the bed's initial characteristics were maintained, with ~50% of the bed regenerated less than five times. This is based on Bayer et al. (2005), who suggested 7–12 refills for an efficient operation of the GAC treatment. Consequently, the total number of bed replacements (N_{BR}) over the lifespan of the unit (60 years) is defined as follows:

$$N_{BR} = T_{treatment} / (n_{max} t_{GAC}) \quad (-) \quad (2)$$

where:

N_{BR} total number of bed replacements over the lifespan of 60 years (–)

T_{treatment} treatment time (days)

n_{max} maximum number of bed regenerations before replacement (10)

t_{GAC} bed service time (days).

The amount of fresh and regenerated GAC was then calculated by adapting the method proposed by Clements (2002):

$$F_{GAC} = m_{GAC} [1 + N_{BR} + m_{loss} (n_r + N_{BR} n_{max})] \quad (kg) \quad (3)$$

$$R_{GAC} = m_{GAC} (n_r + N_{BR} n_{max}) \quad (kg) \quad (4)$$

where:

F_{GAC} amount of fresh GAC needed for the treatment (kg)

m_{GAC} amount of granular activated carbon in the bed (kg)

m_{loss} percentage of GAC lost during regeneration (%)

n_r number of bed regenerations after the previous bed replacement (–)

R_{GAC} amount of regenerated GAC (kg).

The assumptions for the variables in eqns. (1)–(4) are summarised in Table 1. As can be seen, three different EBCTs and bed service times were considered, based on the range of values reported for large GAC treatment facilities (Bayer et al., 2005; Clements, 2002; Creek et al., 2001; Reungoat et al., 2011). The trends for the fresh and regenerated GAC requirements according to the different bed service times are given in Fig. 3; the actual values can be found in Table 1. As expected, the required amount of GAC decreases with increasing the bed service time. These results were used in LCA to determine the influence on the environmental impacts of the variation in the key parameters.

2.2.2.2. Ozonation. As mentioned earlier, the main operating parameters that need to be considered for ozonation units are the

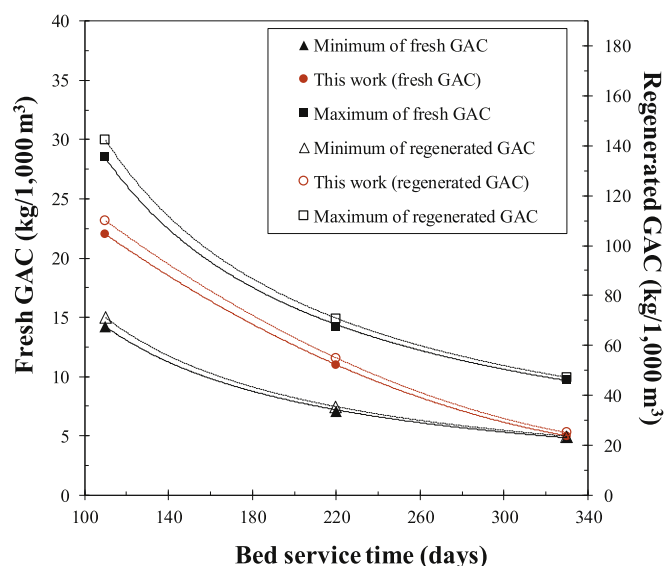


Fig. 3. Estimated ranges of fresh and regenerated granular activated carbon for 1000 m³ of wastewater treated (Black lines: ranges for different bed service times and empty-bed contact times. Red lines: ranges considered in this work. GAC lost during regeneration: m_{loss} = 10%; GAC density: 564 kg/m³). (For interpretation of the references to colour in this figure legend, the reader is referred to the Web version of this article.)

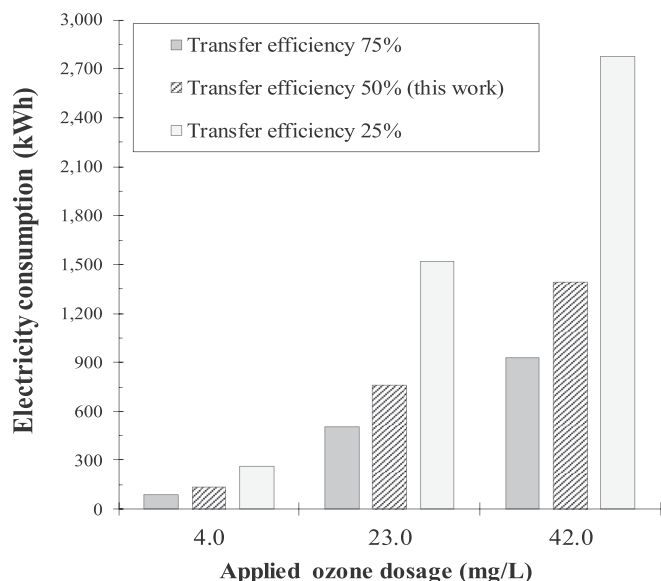


Fig. 4. Estimated electricity consumption per 1000 m³ of wastewater for different ozone transfer efficiencies and applied ozone dosage (Transfer efficiency: 50%).

amount of ozone required for efficient treatment and electricity consumption. The former can be calculated as (Wang et al., 2005):

$$D_{\text{ozone}} = T \frac{100}{\text{TE}} \quad (\text{mg/L}) \quad (5)$$

where:

D_{ozone} applied ozone dosage (mg/L)
 T transferred ozone dosage (mg/L)
 TE ozone transfer efficiency (%).

The range of transferred ozone dosages and the transfer ozone efficiencies considered here can be found in Table 1. These values were chosen to match the ranges found in the literature for achieving higher final water quality from secondary effluents (Wang et al., 2005).

The electricity consumption in the ozonation treatment was estimated as being directly proportional to the amount of wastewater treated and electricity consumption for ozone production, as follows:

$$E_{\text{ozonation}} = D_{\text{ozone}} V_{\text{inf}} E_{\text{ozone}} \quad (\text{kWh}) \quad (6)$$

where:

$E_{\text{ozonation}}$ electricity consumption for ozonation (kWh)
 D_{ozone} applied ozone dosage (kg_{ozone}/m³)
 V_{inf} influent volume to be treated, i.e. the functional unit (m³)
 E_{ozone} electricity consumption for ozone production (kWh/kg_{ozone}).

Based on the data in Table 1, the transferred ozone dosage (T) was estimated in the range from 4 to 42 mg/L (Burns et al., 2007; Petala et al., 2006; Tripathi and Tripathi, 2011; Wang et al., 2005). The average electricity consumption for ozone generation (E_{ozone}) from the air was estimated at 16.5 kWh/kg_{ozone} (Wang et al., 2005). Thus, the estimated electricity consumption for ozonation ($E_{\text{ozonation}}$) ranges from 90 to 2780 kWh/1000 m³ for the transfer efficiency TE between 25% and 75% (Fig. 4). This variation is due to the strong dependence of the TE on the reactor type and size, influent composition and the required level of treatment (disinfection).

2.2.3. Estimation of removal rates of target PPCPs

The concentrations of PPCPs in the effluents of WWTPs vary greatly in the literature, as indicated in Table 2. To account for this variability, the mean concentrations of target PPCPs were considered here (also shown in Table 2), based on the measurements in WWTPs around the globe. A wide variation in the removal efficiencies of advanced treatment systems is also reported (Bellona et al., 2004; Delgado et al., 2012; Gogate & Pandit, 2004a, b; Taber et al., 2009; Yoon et al., 2007; Yu et al., 2008), as they are heavily dependent on the influent composition and operating parameters. Nevertheless, the removal of the PPCPs using the advanced methods considered in this study is commonly reported to be greater than 65%. For this reason, the following range of removal efficiencies is considered: 65%, 75%, 85%, 95% and 99%.

2.2.4. Other inventory data

The inventory data for the construction, operation and decommissioning of the advanced treatment plants over their lifespan are detailed in Table 3; further information can be found in Table S1 in SI. The data for GAC and NF are based on full-scale facilities treating 2000 m³/d (Bonton et al., 2012) while for SPF, data from a pilot-scale study treating 7 m³/d were used (Muñoz, 2006; Muñoz et al., 2007). Hence, it was necessary to scale up the plant to estimate the amount of materials used in the construction of an industrial-size plant. This was carried out by adapting the

Table 2
Concentration of target PPCPs in secondary effluents from WWTP entering the advanced treatment systems.

Compound	Concentration (µg/L)			Sources
	Minimum	Mean	Maximum	
Diclofenac	0.49	0.65	0.80	Tarpani and Azapagic (2018), Verlicchi et al. (2012)
Ibuprofen	0.37	1.99	3.60	Tarpani and Azapagic (2018), Verlicchi et al. (2012)
Trimethoprim	0.12	0.24	0.36	Tarpani and Azapagic (2018), Verlicchi et al. (2012)
Erythromycin	0.73	0.75	0.77	Verlicchi et al. (2012), Tarpani and Azapagic (2018)
Sulfamethoxazole	0.06	0.17	0.28	Tarpani and Azapagic (2018), Verlicchi et al. (2012)
Carbamazepine	0.99	1.02	1.04	Tarpani and Azapagic (2018), Verlicchi et al. (2012)
Estrone	0.03	0.06	0.08	Verlicchi et al. (2012), Tarpani and Azapagic (2018)
17β-Estradiol	0.008	0.009	0.010	Tarpani and Azapagic (2018), Verlicchi et al. (2012)
Triclosan	0.07	0.20	0.32	Tarpani and Azapagic (2018), Verlicchi et al. (2012)

Table 3Inventory data for the advanced effluent treatment plants (per 1000 m³ of wastewater).

Ecoinvent data	Granular activated carbon	Nanofiltration	Solar photo-Fenton	Ozonation	Unit (per 1000 m ³)
Construction and parts replacement					
Steel, low-alloyed	0.0523	0.0063			kg
Reinforcing steel	0.4150	0.0901	1.3947		kg
Glass fibre	0.0175	0.0188			kg
Concrete ^b	0.0008	0.0002			m ³
Polyvinyl chloride		0.1200			kg
Chromium steel 18/8			0.3529		kg
Aluminium, production mix			0.4369		kg
Section bar extrusion, aluminium			0.4369		kg
Anodising, aluminium sheet			0.2521		m ²
Glass tube, borosilicate			0.3025		kg
Operation					
	Activated carbon production ^a (min/mean/max)	Membrane filtration (min/mean/max)	Catalyst (min/mean/max)	Ozone generation (min/mean/max)	
Hard coal supply mix	15/33/66				kg
Hard coal, burned at industrial furnace 1–10 MW	304/669/1338				MJ
Natural gas, burned in industrial furnace >100 kW	66/145/290				MJ
Water, deionized, at plant	60/132/264				kg
Electricity, medium voltage, at grid (Germany)	8.0/17.6/35.2				kWh
Electricity, medium voltage, at grid (UK)		270/412/554		150/750/1300	kWh
Iron sulphate, at plant			13.62/34.06/54.50		kg
	Activated carbon regeneration ^a (min/mean/max)	Cleaning agents (min/mean/max)	Hydrogen peroxide (min/mean/max)		
Hard coal, burned at industrial furnace 1–10 MW	75/165/330				MJ
Natural gas, burned in industrial furnace >100 kW	260/572/1144				MJ
Steam, for chemical process	15/33/66				kg
Electricity, medium voltage, at grid (UK)	0.75/1.65/3.30				kWh
EDTA		0.164/0.250/0.336			kg
Sodium hydroxide, 50% H ₂ O		0.164/0.250/0.336			kg
Hydrogen peroxide, 50% H ₂ O			20/110/200		kg
Other operational data					
Electricity, medium voltage, at grid (UK)	29.0				kWh
Sodium hydroxide, 50% H ₂ O	60.0		80.0	80.0	kg
Aluminum sulphate, powder	80.0				kg
Polymer (acrylonitrile-butadiene-styrene copolymer)	0.30				kg
Carbon dioxide, liquid	14.0	31.0			kg
Calcium hydroxide	7.00	31.0			kg
Phosphoric acid, industrial grade	1.10				kg
Sulphuric acid, liquid		36.0	130		kg
Chlorine, liquid	0.60	0.60			kg
Spiral wound modules ^c		0.3584			kg
Waste management					
Disposal, bitumen, 1.4% water, to sanitary landfill	5/11/22				kg
Disposal, plastics, 15.3% water, municipal incineration		0.3584			kg
Cement, hydrated, 0% water, to residual material landfill			46.06		kg
Decommissioning^d					
Disposal steel, 0% water, to inert material landfill	0.0234	0.0048	0.0697		kg
Disposal, inert waste, 5% water, to material landfill	0.0175	0.0188			kg
Disposal, concrete, 5% water, to inert material landfill	0.4500	0.0900			kg
Disposal, polyethylene, 0.4% water, to sanitary landfill		0.0204			kg
Disposal, aluminium, 0% water, to sanitary landfill			0.0437		kg
Disposal, glass 0% water, to inert material landfill			0.3025		kg
Steel - recycled	0.4439	0.0914	1.3250		kg
Concrete ^b - recycling	1.4250	0.2850			kg
Polyethylene – recycling		0.0996			kg
Aluminium – recycling			0.8301		kg
Chromium steel 18/8 – recycling			0.3352		kg
Transport^e					
Transport lorry, 16–32 t, Euro 5	44/57/81	19.8 ^f	58/80/102	16	t.km

^a Amount of fresh activated carbon (min/mean/max): 5/11/22 kg/1000 m³; amount of regenerated activated carbon over the lifetime of 60 year (min/mean/max): 25/55/110 kg/1000 m³.

^b Concrete density: 2300 kg/m³.

^c See Table S2 in the SI for details.

^d Concrete: 24% recycled and 76% landfilled; glass: 100% landfilled; glass fibre: 100% landfilled; metals: 95% recycled and 5% landfilled; plastics: 83% recycled and 17% landfilled.

^e All distances were set to 200 km except for fresh GAC transport to the wastewater treatment site, assumed at 1000 km (imported from central Germany).

^f Negligible variation.

“economy of scale” relationship, used for the scaling of process plants costs (Coulson et al., 1993), as follows:

$$C_2 = C_1 \left(\frac{C_2}{C_1} \right)^{0.6} \quad (7)$$

where:

C_1 and C_2 material requirements for smaller and larger scale plants, respectively
 c_1 and c_2 respective treatment capacities
 0.6 the “economy of scale” factor.

For ozonation, no construction or decommissioning data were available and these are thus excluded from consideration (see Fig. 1).

Regarding the operating data, for each treatment method, minimum, mean and maximum values were considered for each key parameter discussed in section 2.2.1 (for details, see Table 3). The data for the GAC production and regeneration processes were based on Bayer et al. (2005) and Jeswani et al. (2015). The electricity consumption and the cleaning agents (ethylenediaminetetraacetic acid (EDTA) and sodium hydroxide) used in NF were based on the minimum and maximum values reported in Bonton et al. (2012) and Cyna et al. (2002). For SPF, the dosage of catalyst (iron salts)

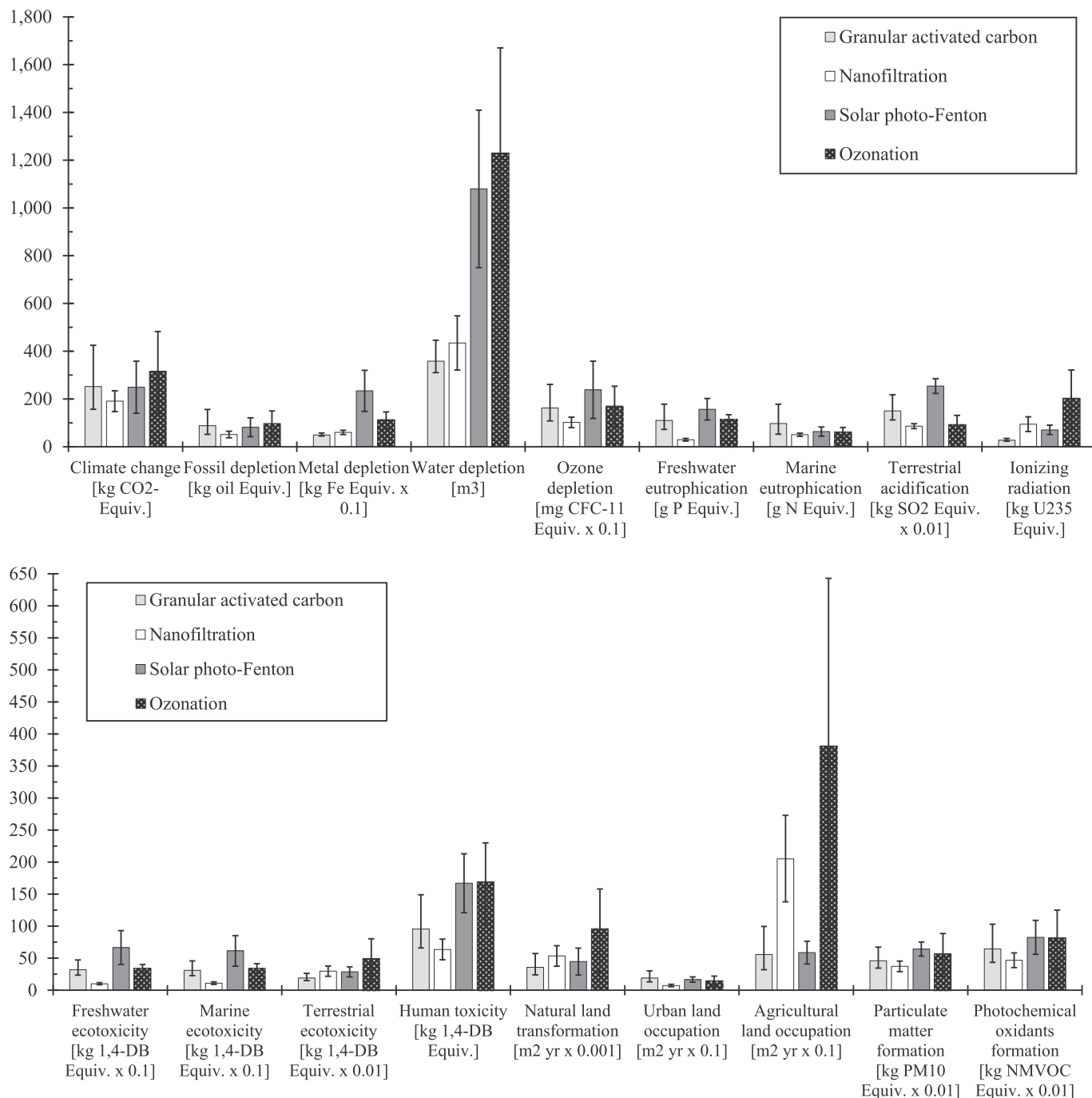


Fig. 5. Life cycle impact of the advanced treatment techniques for PPCP compounds. All impacts are expressed per 1000 m³ of wastewater. The error bars represent the minimum and maximum values for the parameters as specified in Table 3. DB – dichlorobenzene. NMVOC: non-methane volatile organic compounds).

and hydrogen peroxide was based on the studies by [Trovó et al. \(2013\)](#), [Ribeiro et al. \(2015\)](#) and [Klamerth \(2011\)](#). The electricity consumption in SPF was excluded due to a lack of data. Sodium hydroxide used for alkalinity balancing in ozonation was estimated from [Muñoz et al. \(2007\)](#).

The plant parts, such as motors, pipes, pumps, solar panels and ozone diffusers, were assumed to be replaced every 10–15 years. After the end of their useful lifetime of 60 years, the plants were assumed to be decommissioned and waste treated using current waste management practices in the UK for recycling and landfilling of construction materials ([BRE/DEFRA, 2010](#)). The systems were credited for the recycled materials. GAC was assumed to be land-filled after 10 regeneration cycles and NF membranes incinerated at the end of their lifetime of ten years.

2.3. Impact assessment

The ReCiPe 2008 method ([Goedkoop et al., 2009](#)) was used to estimate the environmental impacts of the advanced PPCP treatment options. All eighteen impacts included in ReCiPe are considered here as discussed in the next section. In addition, freshwater ecotoxicity potential of PPCP compounds was estimated using the USEtox methodology ([Henderson et al., 2011](#); [Rosenbaum et al., 2008](#)) to find out if the treatment reduces the ecotoxicity associated with PPCPs on a life cycle basis. Gabi 6.0 ([Thinkstep, 2015](#)) was used for LCA modelling and estimating the impacts.

3. Results and discussion

The LCA results are first presented for the mean operating parameters (see [Table 3](#)) for each impact in turn. The overview of these results is given in [Figs. 5 and 6](#), where the error bars represent the results for the minimum and maximum values of the parameters, discussed in section 2.2.2. As can be seen from [Fig. 6](#), the majority of the impacts across all the treatment techniques are from the operation of the plants.

3.1. Environmental impacts

3.1.1. Climate change potential

The results in [Fig. 5](#) suggest that GAC and SPF have a similar impact on climate change, with the mean values estimated at 252 and 249 kg CO₂ Equiv./1000 m³, respectively. However, the best option is NF with 191 kg CO₂ Equiv. At 316 kg CO₂ Equiv./1000 m³, ozonation is the worst alternative, with around 25% higher impact than GAC. For the latter, 41% of the impact is due to the production of fresh activated carbon and 26% due to the energy used for its regeneration, with the rest being due mostly to aluminium sulphate production. Since fresh GAC is imported from Germany, 4% of the impact is due to road transport.

In NF, 65% of the impact is from electricity generation and the remainder from the productions of liquid carbon dioxide and calcium hydroxide (used for fouling control and effluent balancing). For SPF half of the impact is due to hydrogen peroxide production and another 47% from other operational activities, with the rest being from transport. The majority of the climate change potential of ozonation is from electricity (71%) with the rest being due to sodium hydroxide production.

3.1.2. Resource depletion potential – fossil fuels and metals

As can be seen in [Fig. 5](#), GAC and SPF have similar fossil resource depletion potentials (88 and 81.5 kg oil Equiv./1000 m³, respectively) while ozonation has around 10% higher impact (97 kg oil Equiv.). For GAC, 50% of the impact is related to the production of fresh activated carbon, 24% to energy used for regeneration and 22%

to the treatment process ([Fig. 6](#)). For NF and ozonation, electricity consumption is the main contributor to the depletion of fossil resources (77% and 75%, respectively).

The lowest metal depletion potential was found for GAC and NF (4.9 and 6.0 kg Fe Equiv./1000 m³, respectively) and the highest for SPF (23.4 kg Fe Equiv.). For GAC, the credit for materials recycling after decommissioning reduces the impact by 10% and SPF in 16%. For SPF, the majority of the impact is due to hydrogen peroxide (38%), sodium hydroxide (29%) and sulphuric acid (21%).

3.1.3. Water depletion potential

The highest water consumption was found for ozonation, estimated at 1230 m³ per 1000 m³ of water treated ([Fig. 5](#)). Therefore, more water is consumed along the life cycle than treated. A half of this is due to water consumption during electricity generation and another half from sodium hydroxide production. SPF is the second worst option with 1080 m³/1000 m³. By contrast, GAC and NF consume roughly three times less water.

3.1.4. Ozone depletion potential

NF is the best option for this impact, followed by GAC (10.2 and 16.2 mg CFC-11 Equiv./1000 m³, respectively). The highest impact is from the SPF treatment (23.8 mg CFC-11 Equiv./1000 m³), the majority of which (58%) is due to hydrogen peroxide production. The contribution of transport is also relevant for this treatment option (11%). For ozonation, two thirds of the impact, estimated at 17 mg CFC-11 Equiv./1000 m³, originate from electricity generation and the rest from sodium hydroxide production.

3.1.5. Eutrophication potential – freshwater and marine

SPF has the highest freshwater eutrophication potential, equal to 0.16 kg P Equiv./1000 m³, with the main contributor (~56%) being phosphate emissions to freshwater related to sodium hydroxide production. The equivalent impact for the other treatment alternatives ranges from 0.03 to 0.11 kg P Equiv./1000 m³, almost exclusively due to the operation of the treatment facilities.

GAC has the highest marine eutrophication potential, around 65% higher than the other three methods. Unlike other impacts, the contribution to this impact for GAC is dominated by disposal of activated carbon (57%; see [Fig. 6](#)), due to the organically-bound nitrogen.

3.1.6. Acidification potential – terrestrial

For this impact, SPF is the worst option (2.5 kg SO₂ Equiv./1000 m³), largely due to SO₂ emissions from sulphuric acid production (68%). The next worst option is GAC, with 1.5 kg SO₂ Equiv./1000 m³, of which 83% is from SO₂ emissions from aluminium sulphate production and hard coal burning, used for the production of GAC. NF is the best option at 0.86 kg SO₂ Equiv./1000 m³, followed closely by ozonation at 0.92 kg SO₂ Equiv./1000 m³ ([Fig. 5](#)).

3.1.7. Ionizing radiation potential

As can be seen in [Fig. 5](#), GAC has the lowest ionizing radiation potential (27 kg U235 Equiv./1000 m³). This is almost seven times lower than for ozonation (203 kg U235 Equiv./1000 m³) which represents the worst option for this impact. SPF is the second best option, followed by NF. For all the options, the source of ionizing radiation is nuclear power present in the electricity mix used for the operation of the plant. Given that ozonation is the greatest consumer of electricity among the four options, it is not surprising that it has the highest impact for this category.

3.1.8. Ecotoxicity potential – freshwater, marine and terrestrial

All three types of ecotoxicity exhibit a similar trend, with NF being the best option and SPF and ozonation the worst ([Fig. 5](#)). For

example, freshwater and marine ecotoxicity potentials of SPF are six times higher than those of NF, largely because of emissions to water of copper, nickel and zinc associated with the production of hydrogen peroxide and sodium hydroxide.

For freshwater and marine ecotoxicity, the main contributor to the impacts is the operation of the plants (Fig. 6). However, terrestrial ecotoxicity is largely caused by transport, particularly for GAC and SPF to which it contributes 70%–75%. For ozonation, the

operation of the plant is the main hotspot (85%); transport adds a further 8% to the total impact.

3.1.9. Human toxicity potential

Ozonation has the highest estimated human toxicity potential, equal to 169 kg 1,4-dichlorobenzene (DB) Equiv./1000 m³, 60% of which is from emissions of manganese to freshwaters. The next highest impact is from SPF with 167 kg 1,4-DB-Equiv., with 51%

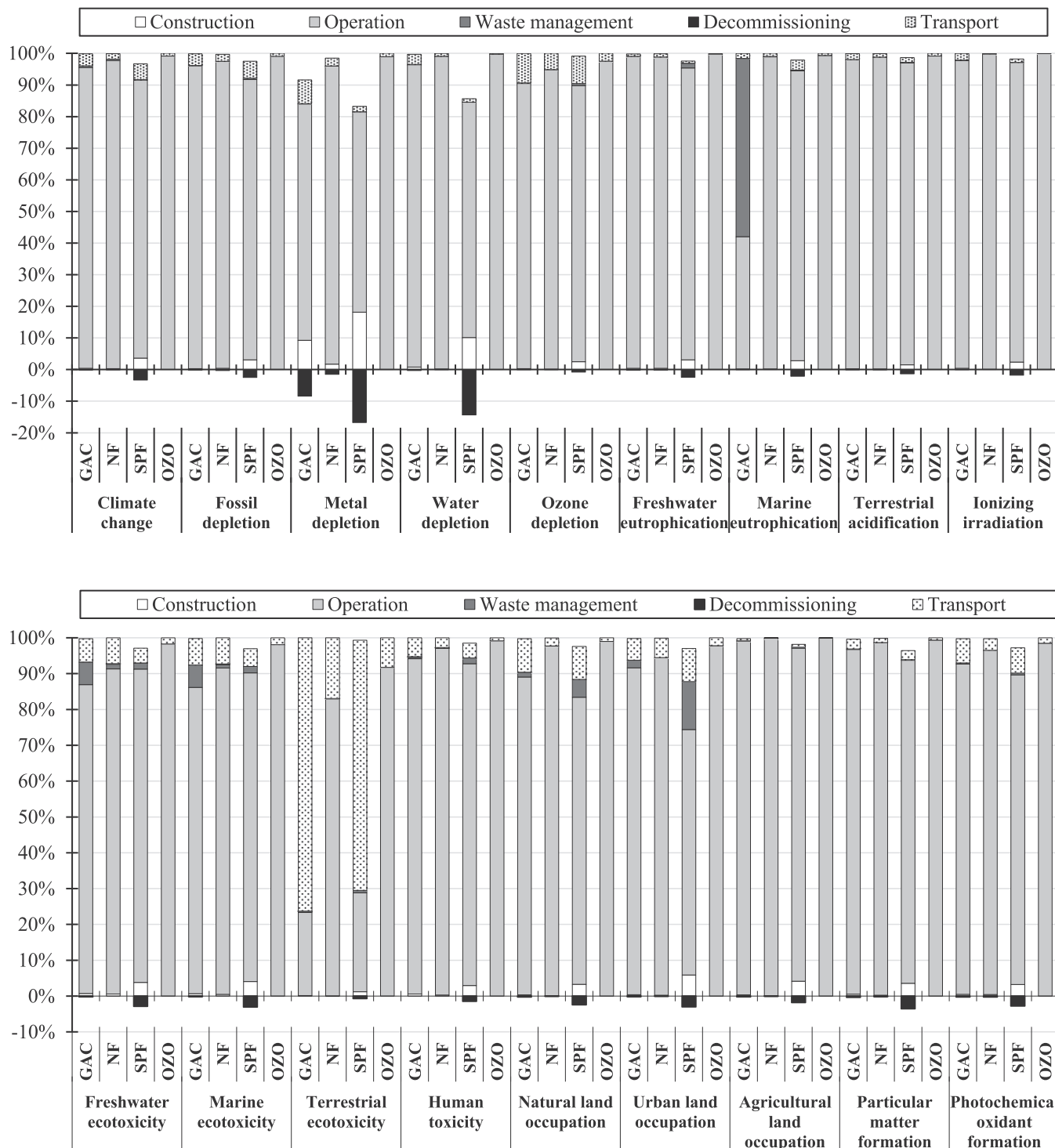


Fig. 6. Contribution of different life cycle stages to the impacts of advanced treatment options (Waste management includes decommissioning).

attributed to the production of sodium hydroxide, again mostly due to manganese emissions. NF is the best treatment alternative for this impact, with a value of 64 kg 1,4-DB-Equiv./1000 m³, followed by GAC at 96 kg 1,4-DB-Equiv.

3.1.10. Land transformation potential – natural, urban and agricultural

Ozonation has the greatest transformation potential for natural and agricultural land, requiring around 0.1 m² yr of natural and 38 m² yr of agricultural land per 1000 m³ of wastewater treated. By contrast, GAC uses 0.04 and 5.6 m² yr, respectively. NF is the best option for urban land and GAC the worst. Much of the land transformation for all options is associated with the operational requirements. For example, for ozonation this is due to forest transformation, industrial areas and landfill sites associated with electricity generation.

3.1.11. Particulate matter formation potential

For this category, NF and GAC are the best alternatives, with a similar impact of ~0.4 kg PM10 Equiv./1000 m³. SPF and ozonation have around a 1/3rd higher impact (~0.6 kg PM10 Equiv./1000 m³). For SPF, the impact is mainly from the productions of sulphuric acid (57%) and sodium hydroxide (19%) due to the emission of SO₂ which contributes to the formation of particulates. For ozonation, emissions to air of NO_x and SO₂ from electricity generation account for 77% of the total potential.

3.1.12. Photochemical oxidants formation potential

At 0.65 and 0.47 kg non-methane volatile organic compounds (NMVOC) Equiv./1000 m³, GAC and NF are the best options for this category. The worst options – SPF and ozonation – have a similar impact (0.82 and 0.81 kg NMVOC Equiv., respectively). The main contributor across all four alternatives is the operation of the plant, related to nitrogen oxide emissions from electricity generation. Transport also contributes around 5%–10% to the impact from GAC and SPF.

3.1.13. Comparison of results with literature

The literature on life cycle environmental impacts of advanced wastewater treatment technologies in general is scarce and, as far as we know, there are no studies that considered specifically the impacts of PPCP advanced treatment. The existing studies also differ greatly in terms of study goals, system boundaries, functional units, assumptions and impact assessment methodologies. Therefore, a meaningful comparison among them is difficult if impossible. Nevertheless, an attempt is made to benchmark the findings of the present study with some others. The focus is on climate change, for which the impact assessment methodology is mainly consistent across different LCA studies. The comparison of other impacts is not possible as other studies did not use the ReCiPe methodology applied here.

For example, [Jeswani et al. \(2015\)](#) considered GAC for removal of organic carbon produced from decaying natural occurring matter. Their estimated climate change potential of 155 kg CO₂ Equiv./1000 m³ agrees well with the lower value in the range obtained here (157–425 kg CO₂ Equiv.). For NF, the only other LCA study found is that by [Bonton et al. \(2012\)](#) which focused on potable rather than wastewater treatment, based in Canada. The reported climate change potential was 40 kg CO₂ Equiv./1000 m³, almost a factor of five lower than the mean value in the present work (191 kg CO₂ Equiv.). In addition to the differences in other assumptions, this discrepancy in the results is probably largely due to Canadian electricity which is largely derived from hydropower, as opposed to fossil fuels prevalent in the UK grid. On the other hand, the impact from SPF estimated by [Muñoz et al. \(2007\)](#) was much higher than

here: 5000 kg vs the mean value of 248 kg CO₂ Equiv./1000 m³. This is likely due to a much higher amount of hydrogen peroxide and catalysts used during the pilot scale experiments to treat wastewater containing methylphenyl glycine.

3.2. Parametric analysis

As mentioned earlier, the results discussed in the previous sections refer to the mean values of the key operating parameters shown in [Table 3](#) for the respective treatment options. To examine the influence of these parameters, a parametric analysis was carried out assuming in turn the minimum and maximum values of the parameters in [Table 3](#). The resulting variations in the impacts are shown as error bars in [Fig. 5](#). As can be seen, most impacts from SPF and ozonation are susceptible to the variations in the key operating parameters and vary widely. Due to this, for some of the categories they become comparable to the other two alternatives. These include climate change, ozone depletion, ecotoxicity and photochemical oxidants, where the minimum values for SPF and ozonation are similar or lower than the respective mean values for GAC. On the other hand, NF showed to be the least sensitive to the variation in the operating parameters.

3.3. Freshwater ecotoxicity potential of PPCP compounds

This section considers freshwater ecotoxicity potential of the target PPCPs when released with the effluent to freshwaters or to agricultural land, the latter if the effluent is used for irrigation. Both the effluents treated by the advanced methods and those not subjected to such treatment but discharged directly after the secondary treatment are considered. The aim is to find out if and by how much the advanced treatment could contribute to reducing the overall ecotoxicity potential of the target PPCPs compared to the effluent from the conventional WWTPs. The USEtox methodology was used for these purposes ([Henderson et al., 2011](#)). Note that the freshwater potential of the advanced treatment options discussed in section 3.1.8 were estimated using the ReCiPe methodology so that the estimates presented in this section are not comparable. Note also that it was not possible to use the ReCiPe methodology to estimate the ecotoxicity potential of the target PPCP compounds due to a lack of their characterisation factors.

The USEtox characterisation factors for freshwater ecotoxicity potential of the target PPCPs are given in [Table 4](#), distinguishing between their potential impact when released to freshwaters and agricultural land. It can be noticed that the latter is much lower than the former for each PPCP.

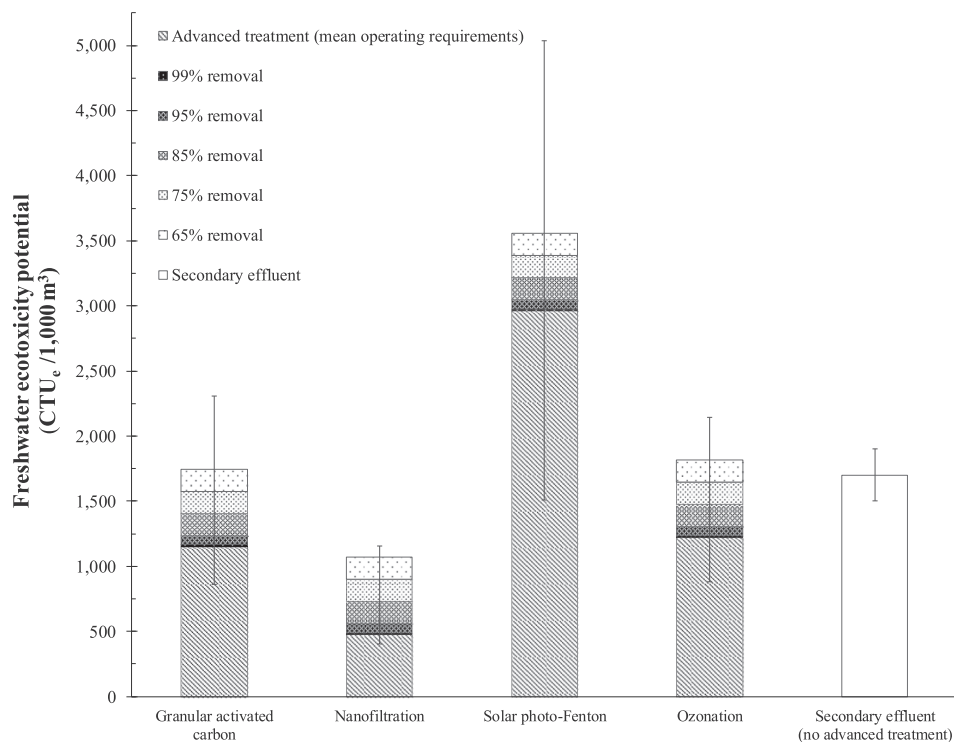
Table 4

USEtox characterisation factors for freshwater ecotoxicity of target PPCP compounds ([Henderson et al., 2011](#)).

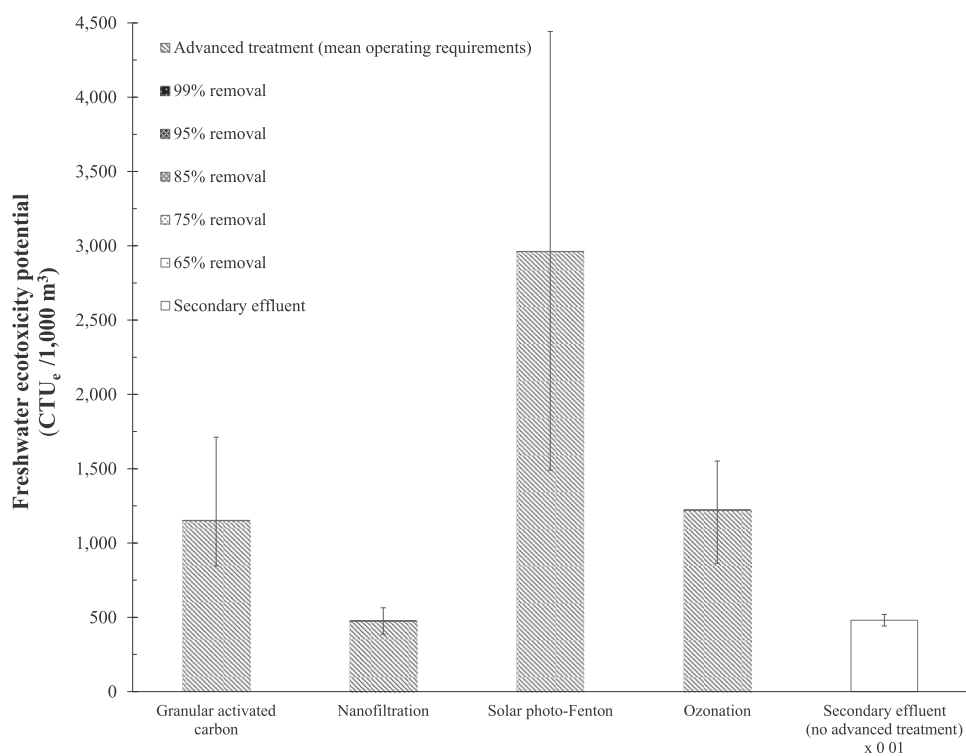
Compound	Characterisation factor (CTU _e /kg) ^a	
	Emission to freshwater ^b	Emission to agricultural land (irrigation) ^b
Diclofenac	2670	105
Ibuprofen	209	4
Trimethoprim	474	19
Erythromycin	24,900	3120
Sulfamethoxazole	2990	195
Carbamazepine	854	13
Estrone	21,400	19
17β-estradiol	184,000,000	255,000
Triclosan	106,000	200

^a CTU_e: comparative toxic units. It represents an estimate of the potentially affected fraction of species (PAF) over time and volume per mass of a compound emitted to the environment. CTU_e/kg = (PAF.m³.day)/kg ([Henderson et al., 2011](#)).

^b CTU_e values from [Alfonsín et al. \(2014\)](#).



a) Release to freshwaters



b) Release to agricultural land

Fig. 7. Freshwater ecotoxicity potential of effluent from advanced treatment plants compared to the impact from the secondary effluent without advanced treatment for different PPCP removal efficiencies (The impact estimated using the USEtox methodology. “Advanced treatment” refers to the life cycle of the treatment process. The impact for “secondary effluent without advanced treatment” in figure b) has been multiplied by 100 as it was too small to show on the graph).

The results are summarised in Fig. 7 and detailed in Tables S3–S6 in the SI. As indicated in Fig. 7a, releasing the PPCPs in the effluent to freshwaters without advanced treatment has the mean ecotoxicity potential of $1700.8 \text{ CTU}_e/1000 \text{ m}^3$ (97% of which is due to 17β -estradiol; see Table S3). By comparison, this is equivalent to the total life cycle freshwater ecotoxicity of GAC treatment at the maximum operating conditions ($1710 \text{ CTU}_e/1000 \text{ m}^3$; Table S4). Fig. 7a also shows that at the mean operating conditions GAC must achieve the removal efficiency above 65% and ozonation must remove at least 75% of the PPCPs to have the ecotoxicity potential comparable to that of the secondary effluent not treated by an advanced method. SPF can have a lower ecotoxicity than the effluents without advanced treatment only if operating at the minimum operating requirements and removing PPCPs with an efficiency of over 85%. The only treatment option capable of reducing the ecotoxicity below that of the secondary effluent without the advanced treatment across the range of operating conditions is NF, with $1157.3 \text{ CTU}_e/1000 \text{ m}^3$ for the maximum operating requirements, mean PPCP concentrations and the lowest removal efficiency (65%). This is 30% lower than the minimum ecotoxicity of the effluent without advanced treatment ($1500.7 \text{ CTU}_e/1000 \text{ m}^3$).

On the other hand, releasing the effluent without advanced treatment to agricultural land achieves a much higher reduction of freshwater ecotoxicity than treating it by any of the advanced treatments. In that case, the mean ecotoxicity potential is equivalent to $4.8 \text{ CTU}_e/1000 \text{ m}^3$ (see Fig. 7b and Table S5 in the SI). This is 354 times lower than the impact of the secondary effluent released to freshwaters for the mean concentrations of PPCPs and 3.5 times lower than the impact when 99% of PPCPs are removed and released to freshwaters (for the latter, see Table S4). The difference between treating and not treating the effluent by an advanced method is even starker when considering its release to land, where the life cycle ecotoxicity of all the treatment options outweighs the impact of the effluent without advanced treatment by several orders of magnitude. For example, NF, the best option in this case, has at least 80 times higher ecotoxicity potential for a 99% removal efficiency ($386 \text{ CTU}_e/1000 \text{ m}^3$; Table S6) than the effluent without advanced treatment released to agricultural land.

Therefore, on the basis of these findings, and also taking into account that the advanced treatment generates many additional impacts as discussed in the previous sections, it could be argued that PPCPs should not be subjected to advanced treatment. Instead, the secondary effluent should be utilised for irrigation of agricultural land rather than released to freshwaters to reduce the overall ecotoxicity. However, if the effluent is released to freshwaters, then nanofiltration should be used as it is the only alternative capable of reducing the ecotoxicity below the impact of the secondary effluent without advanced treatment.

3.4. Wastewater reuse

Agricultural irrigation is at present the most common option for reuse of wastewater in Europe. It is particularly practiced in water-stressed regions. Its use is favoured also because of lower effluent quality requirements compared to potable water, which can be achieved by conventional secondary or simple tertiary treatments (Bdour et al., 2009; Bixio et al., 2006). Furthermore, given that agriculture is one of the largest water consumers, reusing wastewater provides a reliable and cheaper source of freshwater, reduces water stress and the need for other water sources (Barceló and Petrović, 2011; Melin et al., 2006).

As demonstrated in this work, the PPCPs in the effluent discharged after conventional treatment and those subjected to the advanced treatment have low freshwater ecotoxicity potential

when applied to agricultural land, although some compounds, such as diclofenac and carbamazepine, show evidence of soil accumulation (Ternes et al., 2007; Yu et al., 2013; Xu et al., 2009). Still, their removal from wastewaters would potentially create a greater ecotoxicity potential as well as a number of other impacts, such as climate change, acidification, eutrophication, human toxicity, etc.

The results of this work also suggest that the removal of PPCPs to achieve the water quality similar to potable water for release to freshwaters is not environmentally sustainable since it creates a similar or greater freshwater ecotoxicity impact than the effluent without advanced treatment. However, if the treatment is aimed at reuse of treated water for drinking, then advanced effluent treatment is environmentally more sustainable than some drinking-water treatment methods, particularly desalination (Muñoz and Fernández-Alba, 2008; Pasqualino et al., 2011; Sala and Serra, 2004; Shannon et al., 2008). However, many obstacles need to be overcome to enable direct potable reuse of wastewater from advanced treatment methods, such as pumping and buffering requirements (blending ratios with drinking water, reservoir maintenance), economic viability, generation of harmful by-products, regulations and social acceptance (Lim et al., 2008; NRC, 2012; Salgot et al., 2006; Urkiaga et al., 2006).

While advanced treatment of PPCPs may not be warranted, site-control and monitoring of compounds originating from WWTPs effluents and in freshwaters are required to avoid contamination of water sources and consequently drinking water supplies. This is particularly important as water in many cities are found to contain these substances (Benotti et al., 2009; Gaffney et al., 2015; Webb et al., 2003). However, the risks posed to humans by these chemicals in potable water supplies remain unknown (Gaffney et al., 2015; Gibs et al., 2007; Huerta-Fontela et al., 2011; Jones et al., 2005; Kleywegt et al., 2011).

4. Conclusions

This paper considered life cycle environmental impacts of four advanced treatment techniques for nine target PPCP compounds. The results suggest that, on average, NF has the lowest impacts for 13 out of 18 categories. GAC is the best alternative for five impacts, including metal and water depletion, but it has the highest marine eutrophication of all the options. SPF and ozonation are the least sustainable for eight impacts, including acidification and ecotoxicity for the former and climate change and ionizing radiation for the latter. However, most impacts from SPF and ozonation vary widely with the operating parameters and, when considering their ranges rather than the mean values, for some impacts they become comparable to the other two alternatives. These include climate change, ozone depletion, ecotoxicity and photochemical oxidants, where the minimum values for ozonation are lower than the respective mean values for GAC.

On the other hand, GAC and NF are favoured since they have greater removal efficiencies for heavy metals and also avoid production of harmful by-products during the treatment, thus being more suitable for potable reuse of wastewater. Moreover, their life cycle freshwater ecotoxicity is lower than for effluents released from conventional WWTPs to freshwaters without the advanced treatment of PPCPs. GAC, SPF and ozonation should not be used on the grounds of reducing freshwater ecotoxicity.

Overall, releasing the effluent without advanced treatment to agricultural land achieves a much higher reduction of freshwater ecotoxicity than treating it by any of the advanced treatments and releasing to freshwaters. This is due to the freshwater ecotoxicity potential generated by the treatments themselves being higher than the ecotoxicity potential of the PPCPs being treated. Therefore, the use of advanced effluent treatment for agricultural purposes is

not recommended and only NF and possibly GAC should be used if the primary aim is reuse of treated wastewater as potable water.

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Nomenclature

C_1	material requirements for a smaller-scale plant (kg/m^3)
C_2	material requirements for a larger-scale plant (kg/m^3)
c_1	treatment capacity of a smaller-scale plant (m^3)
c_2	treatment capacity of a larger-scale plant (m^3)
D_{ozone}	applied ozone dosage ($\text{kg}_{\text{ozone}}/\text{m}^3$)
EBCT	empty-bed contact time (min)
$E_{\text{ozonation}}$	electricity consumption for ozonation (kWh)
E_{ozone}	electricity consumption for ozone generation (kWh/ kg_{ozone})
F_{GAC}	amount of fresh GAC needed for the treatment (kg)
m_{GAC}	amount of granular activated carbon in the bed (kg)
m_{loss}	percentage of GAC lost during regeneration(%)
N_{BR}	total number of bed replacements over the lifespan of 60 years (–)
n_{max}	maximum number of bed regenerations before replacement (10)
n_r	number of bed regenerations after the previous bed replacement (–)
Q_{inf}	influent flow to be treated (m^3/min)
R_{GAC}	amount of regenerated GAC (kg)
t_{GAC}	bed service time (days)
$T_{\text{treatment}}$	treatment time (days)
V_{GAC}	volume of granular activated carbon in the bed (m^3)
V_{inf}	influent volume to be treated, i.e. the functional unit (m^3)

Appendix A. Supplementary data

Supplementary data related to this article can be found at <https://doi.org/10.1016/j.jenvman.2018.03.047>.

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