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1 **Environmental sustainability of light-driven processes for wastewater treatment**  
2 **applications**

3

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6

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14

15 **Abstract**

16 A comparative analysis is presented of light-driven advanced oxidation processes in  
17 terms of environmental sustainability. Photochemical oxidation has proven a viable  
18 option for treating emerging and priority pollutants at laboratory scale. Nevertheless,  
19 as a nascent technology, photocatalysis is yet to be widely applied at large-scale water  
20 treatment plants. This paper presents a powerful tool that should enable stakeholders to  
21 develop sustainable, large-scale, photocatalytic treatment plants by providing  
22 knowledge of environmental sustainability and hotspots (where technological flaws

23 have high environmental impact) and understanding as to how process sustainability  
24 can be improved through scenario analyses. The following processes were examined:  
25 natural and simulated solar photolysis, solar photo-Fenton without hydrogen peroxide  
26 addition (solar/Fe), solar photo-Fenton (solar/Fe/H<sub>2</sub>O<sub>2</sub>), photolysis under UV-A  
27 irradiation (UV-A), titania-mediated photocatalysis (UV-A/TiO<sub>2</sub>), photolysis under  
28 UV-C irradiation (UV-C), and UV-C treatment with hydrogen peroxide addition (UV-  
29 C/H<sub>2</sub>O<sub>2</sub>). Actual life cycle inventory data were collected at bench scale, and the  
30 environmental performances estimated by means of life cycle assessment. Effective  
31 removal of 1 µg of 17α-ethynylestradiol per liter of wastewater, a commonly occurring  
32 micropollutant and endocrine disrupting chemical, was used as the functional unit.  
33 Solar photolysis exhibited an environmental footprint about 23 times higher than  
34 solar/Fe. Solar/Fe/H<sub>2</sub>O<sub>2</sub> minimized the environmental footprint. Being energy  
35 intensive, simulated solar irradiation had a much higher (~ 5-fold) environmental  
36 footprint than natural solar light. UV photolysis exhibited low environmental impact,  
37 with UV-C found to be about 3 times more environmentally friendly than UV-A  
38 photolysis. Addition of TiO<sub>2</sub> to UV-A and H<sub>2</sub>O<sub>2</sub> to UV-C caused their total  
39 environmental impacts to decrease by about 97% and 88%, implying that UV-A/TiO<sub>2</sub>  
40 was better than UV-C/H<sub>2</sub>O<sub>2</sub>. In terms of total environmental footprint, the advanced  
41 oxidation processes descend in the following order: solar photolysis > UV-A > UV-C  
42 > solar/Fe > UV-A/TiO<sub>2</sub> > UV-C/H<sub>2</sub>O<sub>2</sub> > solar/Fe/H<sub>2</sub>O<sub>2</sub>. The environmental  
43 sustainability of all processes was directly proportional to treatment efficiency but  
44 inversely proportional to treatment time (due to the large energy input per unit time).  
45 Although reagent use (i.e. titania, iron, and hydrogen peroxide) was not associated with  
46 high environmental impact, its addition greatly improved process efficiency as well as  
47 environmental sustainability. For all examined light-driven processes, the main

48 environmental hotspot was electricity consumption. Introduction of renewable energy  
49 sources could reduce the environmental footprint of oxidation processes by up to  
50 87.5%.

51

52 **Keywords:** water purification; estrogens; photocatalysis; LCA; EDCs; EE2

53

54

55

## 56 **1. Introduction**

57 Trace- or micro-pollutants are synthetic chemicals of emerging environmental and  
58 health concern that have recently been detected in the aquatic environment (Tiedeken,  
59 2017). Several hundred endocrine-disrupting chemicals (EDCs) have been measured in  
60 humans and wildlife, even in such remote places as the Arctic (Birnbaum, 2013). There  
61 is growing evidence that these pollutants have adverse effects on human health and  
62 living organisms. Trace-pollutants can act, or have the potential to act, as EDCs that  
63 cumulatively interfere with the endocrine system of living organisms and cause genetic  
64 abnormalities, infertility, feminization, increased cancer rates, trigger Alzheimer  
65 disease, etc. (Rochester, 2013). EDCs derive from the chemical processing industry in  
66 the form of drugs, surfactants, cosmetics, and other personal care products, which  
67 usually end up in the sewage system. Synthetic estrogens are EDCs that are found in  
68 increasing concentrations in natural waters (Zhang et al., 2014) and wastewater  
69 (Mohagheghian et al., 2014). A representative synthetic estrogen is 17 $\alpha$ -  
70 ethynylestradiol (EE2), which is the basic component of the contraceptive pill. EE2 is

71 more stable in an aqueous environment and has greater estrogenic potency (~11–27  
72 times) than natural estrone (E1) and estradiol (E2). Continuous exposure to EE2, even  
73 to concentrations of  $\mu\text{g/L}$ , has been found to cause bodyweight loss, accelerate vaginal  
74 opening, alter estrous cycles in young animals, and damage fish populations (Frontistis  
75 et al., 2015).

76 Due to their xenobiotic and non-biodegradable nature, conventional biological  
77 wastewater treatment plants (WWTPs) cannot effectively remove EDCs, which in turn  
78 are discharged into receiving waters. To overcome this, it is necessary to add robust  
79 tertiary treatment technologies to existing WWTPs. Of the technologies available for  
80 the removal of EDCs, light-driven advanced oxidation processes (AOPs) offer  
81 considerable promise. The effectiveness of AOPs is mainly due to the formation of  
82 reactive oxygen species (ROS), such as hydroxyl radicals ( $\text{HO}^\bullet$ ), which subsequently  
83 oxidize the organic content of water samples. AOPs include solar, UV-A and UV-C  
84 photolysis and photocatalysis, usually accelerated by adding titania ( $\text{TiO}_2$ ) (i.e.  
85 heterogeneous catalysis) (Lee et al., 2017), hydrogen peroxide ( $\text{H}_2\text{O}_2$ ) and/or iron ( $\text{Fe}^{2+}$ )  
86 to form the photo-Fenton reagent (i.e. homogeneous catalysis) (Clarizia et al., 2017).  
87 To date, several studies have investigated the treatment of EE2-contaminated water by  
88 means of UV and solar photocatalysis or photolysis. Marinho et al. (2013) observed  
89 that  $\text{TiO}_2$ -mediated photocatalysis, under solar or UVA irradiation permitted efficient  
90 degradation of EE2, usually at reaction times lower than 15 min. Koutantou et al. (2013)  
91 used a zinc oxide photocatalyst immobilized onto a glass substrate to degrade EE2 by  
92 simulated solar light. They found that at the best conditions assayed, treatment time was  
93 only 50 min. Madsen and Sørensen (2012) found that photocatalysis with  $\text{TiO}_2$  was the  
94 best method for removal of EE2 compared to UVC lamps. Experiments were carried  
95 out in a mobile test unit with wastewater volumes of 30 L. Even so, apart from UV-C

96 photolysis, other light-driven AOPs are still nascent technologies, not yet applied at  
97 industrial-scale.

98 AOPs are energy intensive, with high operating cost and elevated environmental  
99 footprint (Chatzisyneon et al., 2013). Solar photo-Fenton AOPs have high chemical  
100 demand, and generate residual fluxes with negative environmental impacts, such as  
101 sludge contaminated by metal ions, exhausted solid catalysts, etc. (Rodríguez et al.,  
102 2016). Previous research has focused on the degradation efficiency and techno-  
103 economic feasibility of AOPs, without detailed consideration of environmental  
104 sustainability (Rodríguez et al., 2016). A brief review of existing studies on AOPs  
105 environmental sustainability is given by (Ioannou-Ttofa et al., 2017).

106 In order for AOP technology to reach prototype-scale applications, it must be  
107 acceptable from an environmental perspective. To achieve this, the environmental  
108 sustainability of each AOP should first be assessed at bench- or pilot-scale, in order to  
109 identify merits and drawbacks, establish the main environmental impact hotspots, and  
110 assess ways of reducing the total environmental footprint through scenario and  
111 sensitivity analyses. By determining the optimal environmental performance of AOPs,  
112 the technology could be effectively scaled up to sustainable, large-scale applications in  
113 water treatment works.

114 This paper describes a comparative life cycle assessment (LCA) of seven well-  
115 established light-driven AOPs, namely: solar, solar/Fe, solar/Fe/H<sub>2</sub>O<sub>2</sub>, UVA,  
116 UVA/TiO<sub>2</sub>, UVC, and UVC/H<sub>2</sub>O<sub>2</sub>. The aim is to identify the strengths and weaknesses  
117 of AOPs from an environmental sustainability perspective, thus enabling process scale  
118 up. LCA methodology is employed, in accordance with ISO 14040 and ISO 14044  
119 (ISO, 2006a, b), using SimaPro 8. The assessment was made using life cycle inventory

120 (LCI) data collected from bench-scale experiments, rather than extracted from a  
121 database. The results should provide researchers, decision- and policy-makers, and the  
122 water treatment industry with a better understanding of the environmental sustainability  
123 of light-driven AOPs, which in turn should help advance the technology so that it  
124 becomes ready for industrial-scale application. To the best of the authors' knowledge  
125 this is the first study to date dealing with LCA of several light-driven oxidation  
126 processes. Many publications focus on comparing several irradiation sources in terms  
127 of ability to decontaminate/disinfect water and wastewater. Assessment of  
128 environmental sustainability of such processes, including both solar and UV-irradiated  
129 techniques, is presently missing from the literature.

130         Of the various methodologies used to assess the environmental sustainability of  
131 a product or process, the most commonly utilized are multi-criteria analysis (MCA),  
132 environmental performance indicators (EPIs), and life cycle assessment (LCA)  
133 (Hermann et al., 2007). MCA compares and ranks alternative options, and evaluates  
134 environmental consequences according to established criteria. However, its weakness  
135 lies in the subjectivity of the weighting step, necessary to evaluate different criteria.  
136 EPIs estimate the current or past environmental performance of an organisation and  
137 compare it against a set of targets; however, the usefulness of EPIs is limited by  
138 insufficient data availability (Hermann et al., 2007). LCA offers an effective means of  
139 including environmental considerations in the design, production, use, and disposal of  
140 a product (Foteinis et al., 2011). LCA is a tool for the systematic evaluation of  
141 environmental impacts, which provides insight into the overall performance and  
142 relative contributions of different stages within the product lifespan (Hermann et al.,  
143 2007).

144

## 145 **2. Materials and methods**

146 Data used in the comparative LCA analysis were obtained from laboratory experiments,  
147 described by Frontistis et al. (2011, 2012, 2015). All experiments were carried out under  
148 the same ambient temperature and water conditions. Table 1 lists the optimum operating  
149 conditions assayed for each light-driven process. In all cases, the wastewater sample  
150 was stirred by a 50 W magnetic stirrer and the ambient temperature kept constant at  
151  $25\pm 2$  °C. Energy required to keep the temperature constant was external to system  
152 boundaries, while the stirrer was assumed to operate at 30 W (i.e. not at full power). At  
153 industry scale, wastewater pumping would replace the magnetic stirrer. Simulated solar  
154 irradiation was emitted by a Newport, model 96000, 150 W solar simulator system. The  
155 UV-A and UV-C experiments were conducted in an immersion well, batch type,  
156 laboratory-scale photoreactor (Ace Glass, Vineland, NJ, USA). UV-A irradiation was  
157 provided by a 9 W lamp (Radium Ralutec, 9W/78, 350–400 nm). UV-C irradiation  
158 was provided by an 11 W low-pressure mercury lamp (Phillips, TUV PL-S). The  $\text{Fe}^{2+}$   
159 ionic solution used in the experiments was in the form of  $\text{FeSO}_4 \cdot 7\text{H}_2\text{O}$  ( $\geq 99\%$ , Sigma-  
160 Aldrich).  $\text{H}_2\text{SO}_4$  was added in order to regulate the initial water pH.  $\text{TiO}_2$  P25 was  
161 donated by Evonik Industries, and  $\text{H}_2\text{O}_2$  (35% w/w) was purchased from Merck.

162

## 163 **3. Environmental sustainability analysis**

164 To assess the environmental sustainability of light-driven AOPs, LCA methodology  
165 was employed, as detailed in ISO 14040 and 14044 (ISO, 2006a, b). Bench-scale  
166 experimental results were utilized by the environmental model. The timespan covered  
167 2010 to the present date, the geographical boundaries encompassed Greece and similar  
168 countries, and average technology was assumed. For the foreground system, primary



169 inventory data were collected for laboratory-scale experiments, while, for the  
170 background system, data were used regarding the most recent average technology (e.g.  
171 for electricity the average technology mix in Greece was imported from the ecoinvent  
172 database).

173

### 174 **3.1 Functional unit**

175 The functional unit selected to quantify the performance of a light-driven AOP was the  
176 effective removal of 1  $\mu\text{g}$  EE2 per liter of treated wastewater. The life cycle inventory  
177 (LCI) for each AOP under study was then normalized per functional unit (ISO, 2006a,  
178 b) in order to study the environmental performance of the different technologies.  
179 Attributional life cycle assessment (ALCA) was used because it estimates the  
180 environmental impacts of a product or system according to the delivery of a specified  
181 quantity of the functional unit (Chatzisyneon et al., 2016).

### 182 **3.2 System boundaries and life cycle inventory (LCI)**

183 The system boundaries define which unit processes (the smallest elements for which  
184 input and output data are quantified in the LCI) are included within the LCA (ISO,  
185 2006a). Energy and raw material requirements, waterborne emissions, and the  
186 materials' disposal or recycling are included within system boundaries.

187 For the AOPs photoreactor, LCI data could not be identified and so their  
188 primary materials, i.e. glass, lamps, and the stirrer, were taken into account. It was  
189 assumed that solar and UV photoreactors have similar dimensions and materials, and  
190 that all experiments were carried out at the same ambient temperature. Two different  
191 scenarios were examined for the solar AOPs. The first scenario comprised the  
192 photoreactor and lamp (i.e. simulated solar irradiation), whereas the second scenario

193 did not include the lamp (i.e. natural solar irradiation). The latter scenario is closer to  
194 actual operating conditions of solar AOPs. Following Ioannou-Ttofa et al. (2017), the  
195 photoreactor glass was assigned a useful lifespan of five years (10 h/d operation, all  
196 year round). Recycling was also incorporated. Photoreactor lamps are not included in  
197 SimaPro's proprietary life cycle inventory (LCI) databases, and so the LCI data were  
198 obtained from relevant literature (Garrett and Collins, 2009; OSRAM, 2016). The data  
199 were re-scaled according to the power requirements of each process and input to  
200 SimaPro in order to simulate the environmental impact of each lamp under study. Data  
201 on the stirrer used to mix effluent were not available in SimaPro's proprietary LCI  
202 databases, and so were substituted by relevant data concerning the LCI of a low-power  
203 motor (AAB, 2002), re-scaled to fit the rated output of the stirrer under study, and used  
204 as input to SimaPro.

205 Information on the  $\text{Fe}^{2+}$  ion as iron sulphate was supplied from the SimaPro LCI  
206 databases. Residual  $\text{Fe}^{2+}$  in the treated wastewater was also taken into account as  
207 waterborne emission. Data on  $\text{H}_2\text{O}_2$  and  $\text{H}_2\text{SO}_4$  reagents were obtained from proprietary  
208 LCI databases. Energy used to drive each process was supplied as electricity from the  
209 Greek energy grid, which is fossil fuel-dependent and comprises 54% lignite, 11%  
210 crude oil, 17% natural gas, and 18% renewable energy (Ioannou-Ttofa et al., 2017). To  
211 carry out the comparative analysis, from an environmental perspective, of light-driven  
212 AOPs, the final use and disposal route of treated effluent was taken to be external to  
213 system boundaries. In other words, cradle-to-gate (treated effluent) was used.

214

215 Table 1.

216

### 217 **3.3 Life cycle impact assessment (LCIA)**

218 Life cycle impact assessment (LCIA) relates the data inventory to specific  
219 environmental impacts and damages (ISO, 2006a, b). ReCiPe was chosen for the LCIA  
220 as a robust method that comprises both midpoint and endpoint impact/damage  
221 approaches which examine different stages in the cause-effect chain to calculate impact  
222 (Chatzisyneon et al., 2016). The endpoint, or damage-oriented, approach translates  
223 environmental impacts into issues of concern, such as human health, natural  
224 environment, and natural resources. Endpoint results are associated with higher levels  
225 of statistical uncertainty, compared to midpoint, due to data gaps and assumptions  
226 stacking up along the cause-effect chain, but are easier for decision- and policy-makers  
227 to comprehend (Chatzisyneon et al., 2016). Given that this is a comparative LCA,  
228 results are compared using the following three endpoint damage categories: “Human  
229 Health”, “Resources”, and “Ecosystems”. These can be also aggregated into a single  
230 score, which makes interpretation simpler.

231 A hierarchist perspective (H), based on the most common policy principles, was  
232 invoked within ReCiPe along with European normalization and average weighting.  
233 Decisions whether or not to include information in the H model are based on mean  
234 scientific consensus, and it assumes that, with proper management, environmental  
235 impacts can be avoided (Chatzisyneon et al., 2016), thus fitting better the goal and  
236 scope of the comparative analysis.

237 Moreover, in order to ensure accuracy and transparency of the LCA, the primary  
238 LCI data along with data used for the background system were verified against  
239 information from the open literature (Chatzisyneon et al., 2013; Gimenez et al 2015).  
240 Light-driven AOPs comprise a nascent technology for wastewater treatment, and so

241 comparative environmental studies based on similar operating conditions and similar  
242 initial organic loads are needed; however, information on these important parameters is  
243 scarce.

244

### 245 **3.4 Energy consumption**

246 The energy consumption of artificial lighting constitutes a major fraction of the  
247 operating costs in UV treatment. Bolton et al. (2001) introduced the electric energy per  
248 order,  $E_{EO}$ , defined as the energy required for 90% degradation of a pollutant per m<sup>3</sup> of  
249 contaminated water.  $E_{EO}$  (kWh/m<sup>3</sup>/order), for a batch-operated reactor, is calculated  
250 from the following equation:

$$251 \quad E_{EO} = \frac{P \times t \times 1000}{V \times 60 \times \log(C_i/C_f)} \quad (1)$$

252 where  $P$  is the electrical power of the irradiation source (kW),  $t$  is the irradiation time  
253 (min),  $V$  is the volume of the treated effluent (L), and  $C_i$  and  $C_f$  are the initial and the  
254 final pollutant concentrations (mg/L), respectively.

255

## 256 **4 Results and discussion**

257 To render the analysis both comprehensive and straightforward to follow, the results  
258 for the solar and UV irradiation light sources are considered separately. Then, a  
259 comparative analysis of all processes follows in order to identify the most promising  
260 result in terms of environmental sustainability. Finally, a sensitivity analysis is carried  
261 out using scenarios to investigate the effect of the main environmental hotspots and to  
262 propose “greener” alternatives by which to improve sustainability.

#### 263 **4.1 Environmental sustainability of solar-driven AOPs and effects of Fe<sup>2+</sup> and** 264 **H<sub>2</sub>O<sub>2</sub>**

265 Results provided by ReCiPe for natural and simulated solar-driven  
266 photolysis/photocatalysis at endpoint level (Figure 1) show that simulated (artificial  
267 light) and natural solar photolysis yielded by far the highest environmental footprints  
268 of ~11 mPt and ~2 mPt per functional unit, respectively. The environmental footprint  
269 due to photolysis was ~ 23 times larger than that of simulated/natural solar/Fe, using  
270 low reagent concentration (5 mg/L Fe<sup>2+</sup>), with scores of 0.477 mPt (artificial light) and  
271 0.089 mPt (natural light). For photolysis, as well as all other AOPs, the main  
272 environmental hotspot was electricity use derived from Greece's fossil fuel-dependent  
273 electricity mix. At the time of writing, electricity systems worldwide use fossil fuels for  
274 bulk power generation (Berill et al., 2016) and so the foregoing results are presently  
275 valid for Greece, Europe and beyond. Indirect impacts of the use of electricity from  
276 fossil fuels can be traced mainly to the "Human Health" damage category, followed by  
277 "Resources", and less so the "Ecosystem" (Chatzisyneon et al., 2016). "Human  
278 Health" damage is affected by fossil-fuel mining and combustion, which release toxic  
279 materials including metals, sulphur, and polycyclic aromatic hydrocarbons (PAHs) to  
280 the environment (Chatzisyneon et al., 2016). Fossil-fuel extraction and burning  
281 contribute to climate change. Natural gas extraction also releases SO<sub>2</sub>. Impacts from  
282 coal arise from tailpipe emissions after combustion and emissions during blasting at  
283 coal mines (Berill et al., 2016). "Resources" damage is primarily caused by depletion  
284 of fossil fuels for electricity generation and of mineral resources used to construct  
285 equipment required for resource extraction, processing and consumption, and to a lesser  
286 degree by equipment related to AOPs (i.e. the stirrer and photoreactor). Turning to  
287 "Ecosystem" damage, phosphate leachate from coal mining spoil landfill sites and the

288 emission of nitrogen oxides from combustion of fossil-fuel directly impact on  
289 acidification and eutrophication. Waterborne metal emissions from coal power plants,  
290 natural gas extraction (particularly of bromine) and from disposed coal mine spoil  
291 (nickel and magnesium) affect ecotoxicity (Berill et al., 2016; Ioannou-Ttofa et al.,  
292 2017).

293         Use of simulated irradiation raised the environmental impact because the total  
294 environmental footprint of simulated solar photolysis and photocatalysis is about a  
295 factor of 5 higher than natural solar light. This is attributed to electricity consumption  
296 by the lamp (~ 81.3% of total environmental footprint), and to a much lower degree to  
297 the lamp material (~ 0.05% of total environmental footprint). In terms of material, the  
298 stirrer (i.e. motor) contributed 12.4% and 2.3% to the total environmental footprints for  
299 natural and simulated solar photolysis. Finally, the photoreactor material (glass) made  
300 a very low contribution to the total environmental footprint, 0.257% and 0.0494% for  
301 natural and simulated solar photolysis, respectively, mainly because of the long lifespan  
302 of glass whose recycling was included in the system boundaries. The relatively high  
303 environmental footprint of solar photolysis is due to its low treatment efficiency as it  
304 consumes energy during the stirring process while EE2 is removed from wastewater.

305

306 Figure 1.

307

308         To study the environmental impacts of the more environmentally friendly  
309 natural solar-driven AOPs, a separate comparison was undertaken, neglecting  
310 photolysis and simulated solar irradiation. Figure 2 shows that the amount of oxidation  
311 reagents used strongly affected the environmental sustainability of solar-driven AOPs,

312 with high reagent concentration improving the overall environmental sustainability of  
313 solar AOPs. At low concentration of iron ions (5 mg/L  $\text{Fe}^{2+}$ ) the total environmental  
314 footprint of natural solar/Fe was estimated to be 0.089 mPt, whereas when the  
315 concentration was increased to 15 mg/L the total environmental footprint reduced by  
316 about half to 0.047 mPt per functional unit (Figure 2). When  $\text{H}_2\text{O}_2$  was also added as a  
317 reagent, the environmental sustainability of the process was further enhanced. More  
318 specifically, when keeping the iron ion concentration constant at 5 mg/L and adding 10  
319 mg/L  $\text{H}_2\text{O}_2$  the total environmental footprint of the process was  $\sim 0.01$  mPt per  
320 functional unit, and by increasing the  $\text{H}_2\text{O}_2$  concentration to 17.2 mg/L the  
321 environmental footprint of the process achieved a minimum of  $\sim 0.356 \times 10^{-3}$  mPt per  
322 functional unit.

323 This large reduction is attributed to: (a) increased degradation efficiency at  
324 higher  $\text{H}_2\text{O}_2$  concentration (Table 1); (b) lower treatment time (15 min for 10 mg/L  
325  $\text{H}_2\text{O}_2$ , and 1 min for 17.2 mg/L  $\text{H}_2\text{O}_2$ ) and hence reduced energy consumption; and (c)  
326 use of low amounts of  $\text{H}_2\text{O}_2$ , a non-toxic chemical without elevated environmental  
327 impact. As mentioned before, the environmental impacts of solar/Fe can be traced back  
328 to Greece's fossil fuel-dependent electricity mix used to drive the stirrer. The  
329 contributions of electricity consumption to the total environmental footprint of natural  
330 solar/Fe (5 mg/L and 15 mg/L), natural solar/Fe/ $\text{H}_2\text{O}_2$  (10 mg/L) and natural  
331 solar/Fe/ $\text{H}_2\text{O}_2$  (17.2 mg/L) were 87.4%, 87.3% and 86.5%. The photoreactor and the  
332 stirrer-drive motor made material contributions of  $0.256 \pm 0.02$  % and  $12.35 \pm 0.05$  %.  
333 As a non-hazardous reagent when in small concentrations,  $\text{Fe}^{2+}$  had a negligible effect  
334 in all cases (its biggest score was 0.058% in natural solar/Fe/ $\text{H}_2\text{O}_2$  (17.2 mg/L)).  
335 Similarly, the addition of miniscule amounts of  $\text{H}_2\text{SO}_4$  in concentrations of about 50  
336  $\mu\text{L/L}$  led to it also making a negligible contribution. For natural solar/Fe/ $\text{H}_2\text{O}_2$ , addition

337 of hydrogen peroxide at concentrations of 10 mg/L and 17.2 mg/L contributed ~0.037%  
338 and 0.943% to total environmental footprint. The latter, higher percentage contribution  
339 is related to the overall low environmental footprint of the process ( $0.356 \times 10^{-3}$  mPt)  
340 and the higher quantity of hydrogen peroxide used (and the knock-on increased energy  
341 and materials required for its synthesis). It should be noted that no H<sub>2</sub>O<sub>2</sub> emissions (e.g.  
342 airborne, waterborne) or harmful by-products were assumed to be generated during  
343 treatment.

344

345 Figure 2.

346

#### 347 **4.2 Environmental sustainability of UV-A and UV-C photocatalysis**

348 Figure 3 presents the environmental footprints of UV-A and UV-C  
349 photolysis/photocatalysis in terms of “Human Health”, “Resources” and “Ecosystems”  
350 endpoint damage categories. UV-A photolysis yields a higher environmental footprint  
351 (0.309 mPt), whereas that of UV-C is about a factor of three smaller (0.117 mPt). This  
352 is expected because UV-C treatment has a much higher treatment efficiency due to the  
353 higher energy (Frontistis et al., 2015), compared to UV-A treatment. In both cases the  
354 lamp materials hardly contributed to the total environmental footprint, whereas the UV-  
355 C lamp required about 20% higher power but also had significantly higher treatment  
356 efficiency (see Table 1). As a result, UV-C removed 1 µg/L of EE2 at a much faster  
357 rate than UV-A treatment, requiring less energy and contributing less environmental  
358 footprint per functional unit.

359 When reagents were added, the environmental footprint of both UV-A and UV-  
360 C treatment was substantially reduced. Figure 3 shows that addition of titania (10 mg/L



361 TiO<sub>2</sub>) drastically reduced the total environmental footprint of UV-A treatment, from  
362 ~309 μPt for UV-A photolysis to ~9.2 μPt for UV-A/TiO<sub>2</sub> heterogenous photocatalysis.  
363 As far as UV-C treatment is concerned, the addition of H<sub>2</sub>O<sub>2</sub> (10 mg/L) also had a  
364 profound effect, with the environmental footprint of UV-C photolysis reducing from  
365 ~117 μPt for UV-C to ~13.8 μPt for UV-C/H<sub>2</sub>O<sub>2</sub>. These large reductions (~97% for  
366 UV-A/TiO<sub>2</sub> and ~88% for UV-C/H<sub>2</sub>O<sub>2</sub>) are due to a combination of improved treatment  
367 efficiency and reduced treatment time (Table 1).

368 As with solar-driven AOPs, the environmental sustainability of UV-driven  
369 AOPs is enhanced by addition of small amounts of the non-hazardous reagents, TiO<sub>2</sub>  
370 and H<sub>2</sub>O<sub>2</sub>, leading to significant improvement in degradation efficiency and reduction  
371 in treatment time, especially for UV-A treatment.

372 Electricity consumption makes the largest contribution to most damage  
373 categories, reflected by its contribution to the total environmental footprint of UV-driven  
374 AOPs of 88.3 ± 0.1 %. This score is dominated by electricity consumption by the stirrer  
375 motor and, to a lesser degree, to the lamp(s). The stirrer motor as a material was the  
376 next most important environmental hotspot with scores ranging from 9.15% to 9.63%  
377 of the total environmental footprint. The lamp as a material contributed from 1.79% for  
378 UV-A to 2.35% for UV-C. The photoreactor as a material (glass) contributed from  
379 0.19% for UV-C to 0.29% for UV-A. The reagents TiO<sub>2</sub> and H<sub>2</sub>O<sub>2</sub> contributed very low  
380 percentages, 0.236% for UV-A/TiO<sub>2</sub> and 0.0273% for UV-C/H<sub>2</sub>O<sub>2</sub>, of the total  
381 environmental footprint. Even though TiO<sub>2</sub> had a higher impact than H<sub>2</sub>O<sub>2</sub>, UV-C/H<sub>2</sub>O<sub>2</sub>  
382 exhibited a slightly higher total environmental footprint than UV-A/TiO<sub>2</sub>, mainly due  
383 to the reduced treatment time of the latter (Table 1).

384

385 Figure 3.

386

### 387 **4.3 Environmental sustainability of solar versus UV-A and UV-C photocatalysis**

388 Given that photolysis invariably exhibited the highest overall environmental footprint,  
389 the most promising photocatalytic processes were determined in terms of  
390 environmental sustainability. Figure 4 presents a comparative analysis, using ReCiPe  
391 impact assessment method, of natural solar/Fe, natural solar/Fe/H<sub>2</sub>O<sub>2</sub>, UV-A/TiO<sub>2</sub> and  
392 UV-C/H<sub>2</sub>O<sub>2</sub> photocatalysis. Natural solar/Fe/H<sub>2</sub>O<sub>2</sub>, at high reagent concentrations (Fe<sup>2+</sup>  
393 = 5 mg/L and H<sub>2</sub>O<sub>2</sub> = 17.2 mg/L) yielded the lowest score (0.356 μPt per functional  
394 unit) amongst all processes. For simulated solar irradiation, the total environmental  
395 footprint of solar/Fe/H<sub>2</sub>O<sub>2</sub> rose to 1.869 μPt, but nevertheless remains substantially  
396 lower than all the other light-driven AOPs considered. Again, the presence of iron and  
397 hydrogen peroxide oxidants, the reduced treatment time and enhanced EE2 removal  
398 efficiency caused the energy demand per functional unit to be minimized, lowering the  
399 environmental footprint. The next most environmentally friendly AOPs were UV-  
400 A/TiO<sub>2</sub> (~9.2 μPt or ~96% higher than natural solar/Fe/H<sub>2</sub>O<sub>2</sub>) and UV-C/H<sub>2</sub>O<sub>2</sub> (~13.8  
401 μPt). Both exhibited relatively high treatment efficiency, with UV-A/TiO<sub>2</sub> requiring  
402 less treatment time to achieve EE2 removal (Table 1), which meant less energy input  
403 and a lower environmental footprint than UV-C/H<sub>2</sub>O<sub>2</sub>. Also, the lamp required higher  
404 energy to drive the UV-C/H<sub>2</sub>O<sub>2</sub> process (11W) than UV-A/TiO<sub>2</sub> (9 W). The lamps,  
405 photoreactor, and stirrer made low contributions as materials to the total environmental  
406 footprints of the UV-C/H<sub>2</sub>O<sub>2</sub> and UV-A/TiO<sub>2</sub> processes. The contribution by the  
407 reagents, TiO<sub>2</sub> and H<sub>2</sub>O<sub>2</sub>, was miniscule compared to electricity consumption. Finally,  
408 natural solar/Fe exhibited a high overall environmental footprint, especially at low

409 reagent concentration (5 mg/L Fe<sup>2+</sup>) where the value was 0.089 mPt. For a high iron  
410 concentration (i.e. 15 mg/L), the total environmental footprint was halved, to 0.047 mPt  
411 per functional unit (Figures 2 and 4).

412 In short, all the light-driven AOPs considered in this work were limited by the  
413 same environmental hotspot, namely electricity consumption from Greece's fossil fuel-  
414 dependent energy mix, which dominated the contributions to ReCiPe's damage  
415 categories "Human Health" and "Resources". Similar findings were obtained by  
416 (Chatzisyneon et al., 2013) who compared the environmental sustainability of UV-  
417 A/TiO<sub>2</sub> with electrochemical and wet air oxidation processes for treatment of agro-  
418 industrial wastewater.

419

420 Figure 4.

421

422 The present comparison is based on bench-scale experimental data. It is expected that  
423 further benefits can be achieved for all AOPs examined, in terms of lowering the  
424 environmental footprint per functional unit when the processes are scaled up. For  
425 example, in prototype applications, the stirring processes, which required large energy  
426 inputs at bench scale, will be replaced by pumping which is more energy efficient.  
427 Given that it also consumes electricity, pumping is likely to be a prime environmental  
428 hotspot (as also suggested by Foteinis et al. (2018) in a study of pilot-scale Fenton  
429 processes for pharmaceutical wastewater treatment).

430 Energy consumption to degrade 90% of EE2 was also estimated in order to undertake  
431 a more comprehensive comparative analysis of artificial light-driven oxidation  
432 processes. The corresponding treatment time was estimated either using experimental

433 values from photocatalytic tests carried out by Frontistis et al. (2015), Frontistis et al.  
434 (2012) and Frontistis et al. (2011) or by extrapolating the experimental values to  
435 achieve 90% removal of EE2. The results are shown in Table 2, where it is observed  
436 that UVA/TiO<sub>2</sub> process has the lowest energy demands followed by UVC/H<sub>2</sub>O<sub>2</sub>,  
437 solar/Fe(5mg/L)/H<sub>2</sub>O<sub>2</sub>, UVC, UVA, solar/Fe(15mg/L), solar/Fe(5mg/L) and simulated  
438 solar process. In principle, these results are consistent with those obtained from LCA  
439 (Figure 4) confirming the high dependence of AOPs on electricity consumption.

440

441 Table 2.

442

#### 443 **4.5 Sensitivity analysis**

444 The main environmental barrier to light-driven AOPs under study is electricity  
445 consumption from the Greek energy mix dominated by fossil fuels. Power systems  
446 based largely on renewable energy sources (RES) perform much better regarding  
447 climate change and other impact categories than systems based on fossil fuels (Berrill  
448 et al., 2016). A sensitivity analysis was carried out involving three energy mix scenarios  
449 solely based on RES, i.e. solar, wind, and hydropower, all naturally abundant in Greece,  
450 Europe and beyond. Energy storage, curtailment, and grid extension were neglected  
451 because the aim of scenario analysis is purely to illustrate possible pathways and  
452 futures, rather than make forecasts or predictions (Kouloumpis et al., 2015). Moreover,  
453 the extra impacts caused by energy storage and grid extension are likely to be of such  
454 relatively small magnitude that the environmental benefits of switching to renewables  
455 would not be undermined (Berrill et al., 2016). The use of RES to meet the electricity  
456 needs of light-driven AOPs is expected to lead to substantial improvement in their

457 environmental sustainability. For example, use of an electricity mix solely based on  
458 photovoltaic (PV) systems (i.e. 3 kWp single-Si panels mounted on slanted roofs)  
459 decreases the total environmental footprint of solar AOPs by about 85% and UV-driven  
460 AOPs by 87%. On the other hand, use of an electricity mix solely based on wind energy  
461 (onshore wind turbines, capacity in the range from 1 to 3 MW) further improves the  
462 environmental sustainability of light-driven AOPs because energy from wind turbines  
463 usually has a lower environmental impact than solar PVs (Chatzisyneon et al., 2016).  
464 In this case, the total environmental footprint of solar AOPs and UV-driven AOPs is  
465 decreased by about 81% compared to the initial scenario. Finally, use of an electricity  
466 mix solely based on hydropower leads to the largest decrease in total environmental  
467 footprint of light-driven AOPs by 86% (solar) and 87% (UV) because hydropower is  
468 the most environmentally friendly RES option (Ioannou-Ttofa et al., 2017).

469 In all cases, the highest reduction in environmental footprint occurred for the  
470 most energy intensive AOPs (i.e. simulated solar, UV-A, and UV-C, ordered from  
471 higher to lower reduction), whereas the smallest reduction occurred for the most energy  
472 efficient AOPs (i.e. solar/Fe/H<sub>2</sub>O<sub>2</sub>, solar/Fe, UV-A/TiO<sub>2</sub>, and UV-C/H<sub>2</sub>O<sub>2</sub>, ordered  
473 from lower to higher reduction). The order of light-driven processes in terms of  
474 environmental sustainability remained the same for all scenarios; from higher to lower  
475 score: natural or simulated solar > UV-A > UV-C > natural or simulated solar/Fe > UV-  
476 A/TiO<sub>2</sub> > UV-C/H<sub>2</sub>O<sub>2</sub> > natural or simulated solar/Fe/H<sub>2</sub>O<sub>2</sub>. Even so, it should be noted  
477 that UV-A/TiO<sub>2</sub> and UV-C/H<sub>2</sub>O<sub>2</sub> exhibited similar environmental footprints when  
478 using RES.

479

## 480 **5. Conclusions**

481 This paper has investigated the environmental performance of light-driven AOPs at  
482 removing an endocrine disruptor, EE2, from wastewater using actual life cycle  
483 inventory (LCI) data. It was found that the environmental sustainability of light-driven  
484 AOPs was directly proportional to treatment efficiency (which was expected given that  
485 the chosen functional unit was the removal of 1 µg EE2 per liter of wastewater), and  
486 was also inversely proportional to treatment time. Moreover, electricity consumption  
487 from the fossil fuel-dependent Greek energy mix was the main environmental hotspot  
488 for all examined AOPs. The Fe<sup>2+</sup>, H<sub>2</sub>O<sub>2</sub>, and H<sub>2</sub>SO<sub>4</sub> reagents used in light-driven AOPs  
489 were associated with low environmental impacts because the chemicals did not  
490 detrimentally affect health or the eco-system, no harmful by-products were generated,  
491 and only low dosages were used. Use of RES to meet the electricity needs of light-  
492 driven AOPs substantially improved their environmental sustainability, by up to 87%  
493 for solar- and 88% for UV-driven AOPs.

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590 **List of Tables**

591 Table 1: Experimental data, taken from Frontistis et al. (2011, 2012, 2015), used to  
592 build the LCI of light-driven AOPs.

593 Table 2: Electrical energy ( $E_{EO}$ ) consumed by several light-driven oxidation processes.

594 Table 1.

Light-driven processes	Irradiation power, W	[TiO <sub>2</sub> ], mg/L	Power for water stirring, W	[Fe <sup>2+</sup> ], mg/L	H <sub>2</sub> O <sub>2</sub> , mg/L	Treatment time, min	EE2 removal, µg/L	Reference
Solar	150	-	30	-	-	60	2	(Frontistis et al., 2015)
Solar/Fe	150	-	30	5	-	60	46	(Frontistis et al., 2015)
	150	-	30	15	-	60	86	(Frontistis et al., 2015)
Solar/Fe/H <sub>2</sub> O <sub>2</sub>	150	-	30	5	10	15	98	(Frontistis et al., 2015)
	150	-	30	5	17.2	1	196	(Frontistis et al., 2011)
UVA	9	-	30	-	-	60	17	(Frontistis et al., 2015)
UVA/TiO <sub>2</sub>	9	750	30	-	-	10	95	(Frontistis et al., 2012)
UVC	11	-	30	-	-	60	47	(Frontistis et al., 2015)
UVC/H <sub>2</sub> O <sub>2</sub>	11	-	30	-	10	15	100	(Frontistis et al., 2015)

595

596

597 Table 2.

Light-driven processes	Irradiation power, kW	Volume, L	Treatment time to remove 90% of EE2, min	$E_{EO}$ , kWh/m <sup>3</sup> /order	Reference
Solar	0.150	0.3	2251	18758	(Frontistis et al., 2015)
Solar/Fe(5mg/L)	0.150	0.3	115	958	(Frontistis et al., 2015)
Solar/Fe(15mg/L)	0.150	0.3	70	583	(Frontistis et al., 2015)
Solar/Fe(5mg/L)/H <sub>2</sub> O <sub>2</sub>	0.150	0.3	2	17	(Frontistis et al., 2015)
UVA	0.009	0.3	312	156	(Frontistis et al., 2015)
UVA/TiO <sub>2</sub>	0.009	0.3	7	4	(Frontistis et al., 2012)
UVC	0.011	0.3	113	69	(Frontistis et al., 2015)
UVC/H <sub>2</sub> O <sub>2</sub>	0.011	0.3	10	6	(Frontistis et al., 2015)

598

599

600 **List of Figures**

601 Figure 1. Environmental footprint of natural and simulated solar (a) photolysis and (b)  
602 photocatalysis per functional unit, i.e. removal of 1 µg EE2 per liter of wastewater.

603 Figure 2. Environmental footprint of natural solar photocatalysis for removal of 1 µg  
604 EE2 per liter of wastewater. Inset: environmental footprint of natural solar/Fe/H<sub>2</sub>O<sub>2</sub>  
605 (17.2 mg/L).

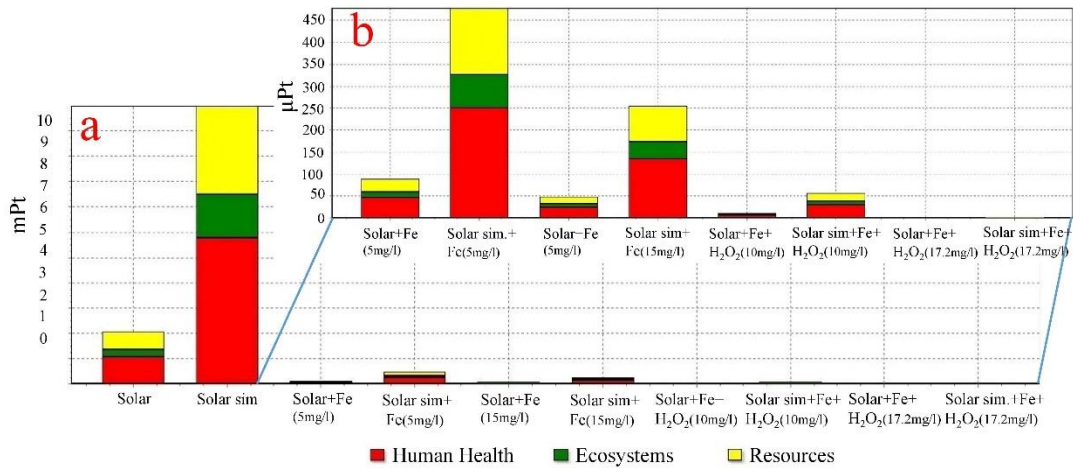
606 Figure 3. Environmental footprint of UV-A and UV-C AOPs per functional unit  
607 (removal of 1 µg EE2 per liter of wastewater).

608 Figure 4. Environmental footprint of natural solar, UV-A and UV-C photocatalysis for  
609 removal of 1 µg EE2 per liter of wastewater.

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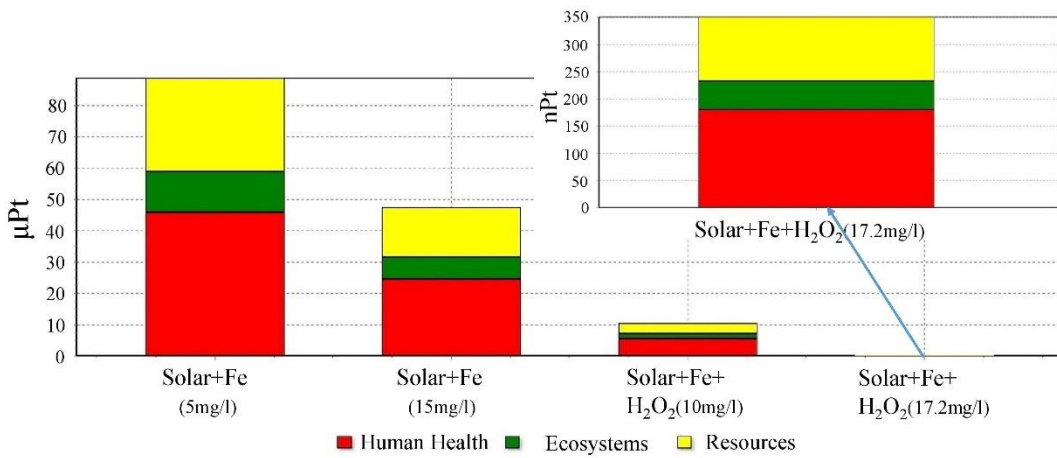


Method: ReCiPe Endpoint (H) V1.10/Europe ReCiPe II/A/Single score

613

614 Figure 1.

615



Method: ReCiPe Endpoint (H) V1.10/Europe ReCiPe H/A/Single score

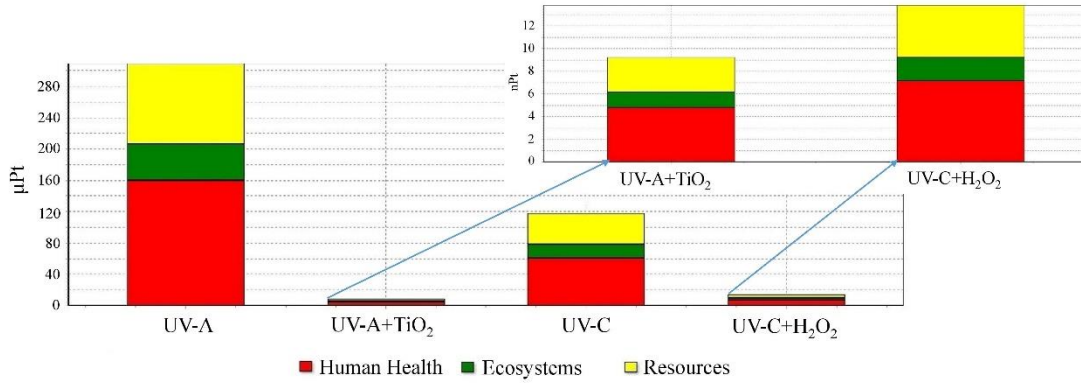
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617 Figure 2.

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Method: ReCiPe Endpoint (H) V1.10/Europe ReCiPe H/A/Single score

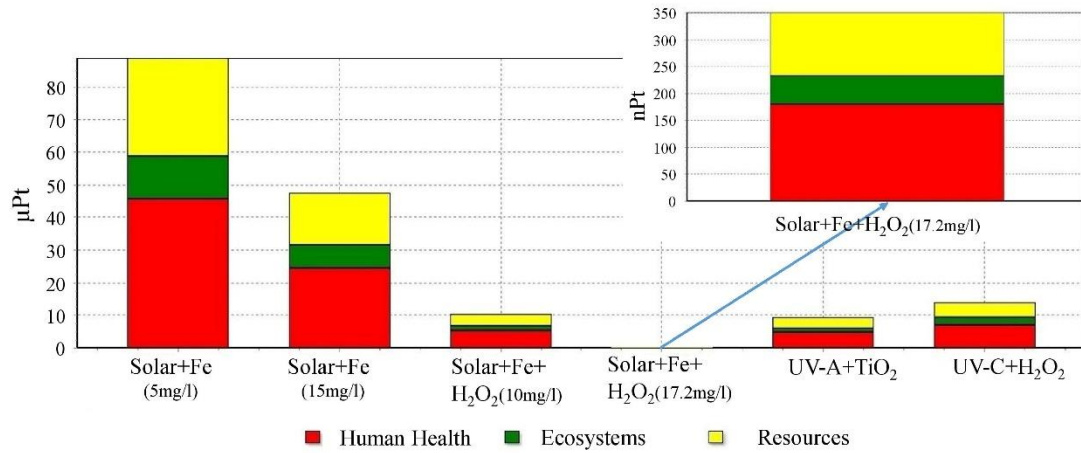
621

622 Figure 3.

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Method: ReCiPe Endpoint (H) V1.10/Europe ReCiPe H/A/Single score

626

627 Figure 4.

628