

Radiation collector systems comparison in Contaminants of Emerging Concern degradation by solar heterogeneous photocatalysis

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Research Article

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1 **Title page**

2 Radiation collector systems comparison in Contaminants of Emerging Concern degradation by solar
3 heterogeneous photocatalysis

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8

9 **Abstract**

10 Wastewater with Contaminants of Emerging Concern (CEC) can be generated from different
11 sources as industry, agriculture and urban and hospital wastes. Heterogeneous Photocatalysis (HP) with
12 TiO_2 is one of the Advanced Oxidation Processes (AOPs) most suitable for water treatment with CEC. In
13 this research, three CEC: Safranin T (SF), 2,4-dichlorophenoxyacetic acid (2,4-D) and Sulfacetamide
14 (SAM) degradation was evaluated by solar-HP in a quartz wall reactor. First, 365 nm wavelength
15 radiation was used and the best operating conditions was determined under the high flow and aeration
16 configuration, obtaining a removal rate of 48.05% for SF, 11.64% for 2,4-D and 6.98 for SAM. Then,
17 under these conditions, SF, SAM and 2,4-D degradation with solar lighting was made on 4 radiation
18 collector systems configurations, Flat Plate Collector (FPC), V Collector (VC), Parabolic Collector (PC)
19 and Compound Parabolic Cylinder Collector (CPC) until reaching the same value of accumulated energy
20 (122.77 kJ m^{-2}) finding that the PC had the best performance in the treatment for the three pollutants.
21 Finally, the Collector Impact Ratio Factor (CIRF) for the pollutants was calculated, achieving until 12
22 times degradation for SAM.

23

24 **Keywords**

25 Contaminants of Emerging Concern, Advanced oxidation processes, Radiation collectors, TiO_2 ,
26 Solar radiation, Safranin, 2,4-dichlorophenoxyacetic acid, Sulfacetamide.

27 **Declarations**

28 **Ethics approval and consent to participate:** Not applicable.

29 **Consent for publications:** Not applicable.

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31 study are available from the corresponding author on reasonable request.

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36 **Authors' contributions:**

37 Fidel Granda-Ramírez: Designed and conducted the experiments, analyzed, interpreted data and was a
38 major contributor in writing the manuscript.

39 Melissa Barrera: Designed and conducted the experiments, analyzed, interpreted data and helped writing
40 the manuscript.

41 Sara Castrillón: Designed and conducted the experiments, analyzed, interpreted data and helped writing
42 the manuscript.

43 Lady Rueda: Designed and conducted the experiments, analyzed, interpreted data and helped writing the
44 manuscript.

45 Juan Pino-Arango: Conducted the experiments.

46 Gina Hincapié-Mejía: Check the manuscript.

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49

50

51 **1. Introduction**

52 Currently, there is a growing interest in Contaminants of Emerging Concern (CEC) whose
53 presence in the environment has largely gone unnoticed in terms of distribution and / or concentration
54 (Tanga 2020). These involve a wide range of chemical compounds that are used in daily life, including
55 medicines, personal hygiene products, soaps, surfactants, industrial additives, plasticizers, pesticides and
56 a wide variety of chemical compounds that are present in the environment (Rodil 2019). Many of these
57 pollutants are not eliminated through conventional systems; and, for the most part, correspond to
58 substances not regulated by the environmental authorities. Among the dyes belonging to the CEC group,
59 is Safranin T (SF), which is a biological dye that is frequently used to dye tissues, the detection of
60 structures in eukaryotic and prokaryotic cells and also, it's most common use it is in Gram staining
61 (Aguirre 2012). 2,4-Dichlorophenoxyacetic Acid is one of the most widely used compounds in
62 commercial herbicides, being responsible for causing devastating effects on the environment, especially
63 in aquatic ecosystems and human health. Due to its persistent nature, it is not degraded efficiently by
64 wastewater treatment plants, and it is necessary to use other more effective degradation methods such as
65 Advanced Oxidation Processes (Samir 2015). On the other hand, Sodic Sulfacetamide (SAM) is an
66 ophthalmic drops or ointment in the treatment of eye infections, it is provided orally for the treatment of
67 acne and seborrheic dermatitis, and is rapidly excreted in the urine and it can be found in soap, shampoo,
68 cream and washing solutions (Bendjama 2018).

69

70 Advanced Oxidation Processes (AOP) are defined as effective methods for the removal of a
71 large number of persistent contaminants. These make changes in the chemical structure of pollutants until
72 they reach mineralization, transforming organic matter into carbon dioxide and water (Samir 2015). One
73 of the most AOP used is the Heterogeneous Photocatalysis (HP) in which a semiconductor is irradiated to
74 generate reactive species and the greatest used in photocatalytic applications is TiO₂, particularly the P-25
75 (Evonik®), because it presented a higher photocatalytic activity, is not toxic, is stable in aqueous
76 solutions and It is not expensive (Bendjama 2018). In photocatalytic process with TiO₂ in aerobic
77 environments, when ultraviolet light radiation striking a surface of a semiconductor, this one is capable of
78 generating electron-hole pairs and Reactive Oxygen Species (ROS) such as hydroxyl radicals ($\bullet\text{OH}$),
79 superoxide anions ($\bullet\text{O}_2^-$), hyperoxide radicals ($\bullet\text{OH}_2$) or Hydrogen Peroxide (H₂O₂) (Kanakaraju 2018). In

80 photocatalytic processes with TiO₂, in addition to properly locating the catalyst (either dispersed or
81 through the implementation of supports) it is necessary to achieve an efficient exposure to the useful light,
82 to ensure an adequate process, said exposure can be carried out using systems of radiation collectors.
83 Globally, various collector designs have been proposed which seek to increase efficiency and reduce the
84 costs of photocatalytic systems for decontamination and water treatment (Zhu 2019). HP with TiO₂ as a
85 catalyst uses only the UV fraction of the solar spectrum (direct or diffuse). An important aspect of the
86 collectors is the refractive surface that covers them, since this has the purpose of directing and reflecting
87 the useful light towards the reactor to achieve maximum use of it and avoid unnecessary losses. Among
88 the best reflective materials are aluminum-based mirrors (Blanco 2004).

89 An important aspect of the collectors is the refractive surface that covers them, since this has the
90 purpose of directing and reflecting the useful light towards the reactor to achieve maximum use of it and
91 avoid unnecessary losses. Among the best reflective materials are aluminum-based mirrors (Bandala
92 2004). The most widely used collector designs are Parabolic Collectors (PC) (Figure 1), V (VC) (Figure
93 2) and Compound Parabolic Cylinder (CPC) (Figure 3) among others (Bandala 2004).

94

95 **Figure 1.** Parabolic Collector (PC).

96 **Figure 2.** V Collector (VC).

97 **Figure 3.** Compound Parabolic Cylinder Collector (CPC).

98

99 Due to the problem that has triggered the generation of CEC, in recent years, proposals have
100 been proposed for the treatment of these compounds through the AOP. Collectors without radiation
101 concentration have been widely used in the HP, the CPC system being one of the most efficient and with
102 the best technology available (Olleros 2013). The HP implementation with TiO₂ using sunlight as a
103 radiation medium and using a CPC as a collector has proven to be an adequate system for the degradation
104 of a mixture of 15 CEC, obtaining degradation of 90% of the pollutants (Maldonado).

105 Through this research, a comparison of 4 radiation collectors configurations Flat Plate Collector
106 (FPC), V Collector (VC), Parabolic Collector (PC) and Compound Parabolic Cylinder Collector (CPC)

107 covered with a reflective material was performed, in which the most efficient collection system for SF,
108 2,4-D and SAM degradation using solar radiation was determined.

109

110 2. Materials and methods

111 In this research were used TiO₂ P-25 (Evonik®), Safranin T (Carlo Erba, 100%), as a source of
112 2,4-dichlorophenoxyacetic acid, a commercial herbicide PROFIAMINA® 720 SL was used with a
113 content of 720 g L⁻¹ thereof and Sodium sulfacetamide was provided by Corpaul (Medellín, Colombia). In
114 the first stage of this research, a 1 liter reservoir was used to contain the solution to be treated; this
115 solution is recirculated by an immersion pump (JAD Reference FP-750) to the cylindrical reactor with
116 quartz walls (internal diameter 2.0 cm, external diameter 2.2 cm and length of 13 cm). This reactor has an
117 effective irradiation volume of 35.5 cm³. The system was equipped with an UV lamp (MoodLites,
118 maximum emission wavelength at 365 nm and 13 W of power) to determine the best reaction conditions
119 and thus proceed to degrade the CEC with solar irradiation. For all tests, 500 mL of a solution at
120 20 mg L⁻¹ of the pollutants was prepared, corresponding to the amounts found in previous studies
121 (Granda-Ramírez 2017). Initially, for determination of the best degradation conditions using UV lamp, SF
122 was used. Subsequently, the high and low flow rates were determined by averaging nine measurements,
123 obtaining the values of 94.84 mL s⁻¹ and 30.73 mL s⁻¹. Likewise, the effect of the presence of oxygen in
124 the system through an air supply was evaluated with an aerator pump (Jeneca Reference AP-9800 with a
125 flow at 1.6 L min⁻¹). Variations in flow rate and aeration (Table 1) were made with the objective of
126 finding a reactor operating condition that allows the best possible degradation of the contaminant. In the
127 follow of pollutants degradation a spectrophotometer Jenway – 7200 series was used. The SF was
128 measured at 520 nm, 2,4-D at 280 nm and SAM at 260 nm (Figure 4).

129 **Table 1.** Experimental design to determinate the best operation conditions..

Exp.	Air	Q (mL s ⁻¹)	Rad	TiO ₂
C1	NO	High		X
C2	NO	Low	X	
C3	NO	High	X	
E1	NO	Low	X	X
E2	NO	High	X	X
E3	YES	Low	X	X
E4	YES	High	X	X

130

Figure 4. First stage's reaction system.

TiO₂ was dispersed in the system with a concentration of 1 g L⁻¹ (Granda-Ramírez 2017). A dark absorption test was initially applied to establish the affinity between safranin and the catalyst, additionally (C1, Table 1), evaluation of the pollutant photolysis operating at high and low flow rates was made (C2 and C3, Table 1). The HP was evaluated by varying the flow rate and the air presence as described in Table 1 (E1 – E4). Once the best reaction conditions had been established in the first stage, the treatments of the contaminated waters with SF, SAM and 2,4-D (20 mg L⁻¹) were carried out under solar irradiation using 4 radiation Collectors configurations FPC, VC, PC, CPC coated by an aluminum reflective material (second stage). The comparison of the pollutants degradation was made until to accumulate 122.77 kJ m⁻² of energy in the system. Collectors designed were made through 3D modeling with SketchUp® software and the elaboration was done with a Fused Form 600 ® brand 3D filament printer, using a PLA filament.

3. Results and discussion

3.1 First stage: determination of best degradations conditions.

For the first stage, Figure 5 shows that SF Dark adsorption showed a 9.8% of contaminant adsorbed in the catalyst, establishing that there is an affinity between SF and TiO₂ that favors its degradation. Through the photolysis experiments, a higher degradation of SF at a high flow rate was determined obtaining a removal of 8.68%, while a 2.98% at a low flow rate was obtained. According to the results obtained it was determined that the oxygen presence favors the degradation of the contaminant. On the other hand, it was determined that the high flow rate (94.84 mL s⁻¹) gives a greater degradation of the contaminant. This flow has a speed 3 times higher compared to the low flow (30.73 mL s⁻¹), this indicates that the solution to be treated experiences a longer contact time with the radiation which allows a greater degradation and efficiency of the system. Analyzing the flow and aeration parameters, the best operating condition for degradation was provided by the E4 (Table 1) experiment (48.05%) set by a high flow rate and air supply.

158 **Figure 5.** First stage results to determine the best conditions for SF degradation.

159 In this way, it is determined that the air supply present in experiments E3 and E4 favors the
160 degradation of the pollutant. Since the air present in the system allows oxygen to react with the excited
161 electron, forming the radical superoxide anion ($\bullet\text{O}_2^-$) and other ROS contributes to the oxidation of the
162 pollutant (Doménech 2004). On the other hand, it was possible to observe that the high flow rate (94.84
163 mL s^{-1}) grants greater pollutant degradation as evidenced in experiments E2, E4 and in high flow
164 photolysis. This flow has a speed 3 times higher compared to the low flow (30.73 mL s^{-1}), this indicates
165 that the solution to be treated experiences a longer contact time with radiation, which allows better
166 degradation and operation of the system.

167 Analyzing the flow and aeration parameters, the best operating condition for degradation was
168 provided by experiment E4 (48.05%) configured by a high flow rate and air supply (Figure 5). With the
169 previous results, the other pollutants selected for this study (2,4-D and SAM) were degraded. Figure 6
170 displays the 3 pollutants degradation rates using the optimal conditions found for SF, showing the
171 resistance that some molecules show to being degraded.

172

173 **Figure 6.** CEC degradation in best conditions using a UV lamp without collector.

174

175 **3.2 Radiation collectors design.**

176 Collectors design was made based on the reactor internal (2.1 cm) and external (2.2 cm) diameter
177 and its length (13 cm). For each of the collectors, except for the plane, the corresponding design equations
178 used for each geometry will be presented below.

179 For V Collector (VC), its design was based on the absolute value function (Equation 1) (Bandala
180 2004), with values for x between -3 and 3, which provides an angle of 90 degrees, it was possible to
181 design the sensor through 3D modeling (SketchUp®) as shown in Figure 7.

182
$$f(x) = |x| \tag{1}$$

183 **Figure 7.** V Collector. a) 3D SketchUp® model. b) 3D printed VC.

184 On the other hand, for the parabolic collector (PC) with the reactor measurements and based on
185 the parabola function (Equation 2) (Blanco 2004), Equation 3 is obtained, with which the parabolic
186 collector 3D modeling (Figure 8a) and subsequently 3D printed (Figure 8b).

$$187 \quad (h; k)^2 = 4p(y - k) \quad (2)$$

188 Where:

189 (h; k) is the vertex with coordinates (0; 0)

190 p is the focal length (2.2 cm)

191 (h; k + p) is the focus with coordinates (0; 2.2)

$$192 \quad f(x) = \frac{x^2}{8.8} \quad (3)$$

193 **Figure 8.** Parabolic Collector. a) 3D SketchUp® model. b) 3D printed PC.

194

195 Knowing that a Compound Parabolic Cylinder Collector (CPC) is going to be designed with a
196 concentration factor equal to 1 and that the photoreactor's external radius is 11 mm, when using the
197 radiation concentration equation (Equation 4), it was determined that the CPC opening (a) is 69.11mm
198 (Blanco 2004).

$$199 \quad CR = \frac{a}{2*\pi*r} \quad (4)$$

200 Where:

201 CR: concentration factor.

202 a: collector opening (mm).

203 r: reactor external radius (mm).

204 Then, based on the involute equation (Equation 5) and with the reactor measurements, equation 6
205 is obtained, which corresponds to the reactor involute that would be one of the CPC sheets (Blanco 2004).

$$206 \quad f(\theta) = DIR + RI(\theta) \quad (5)$$

207 Where:

208 DIR: reactor internal diameter (mm)

209 RI: reactor internal radius (mm)

$$210 \quad f(\theta) = 21 + 10.5 (\theta) \quad (6)$$

211 Finally, in order to print the CPC, it is necessary to transform equation 6 to rectangular
212 coordinates (Equation 7).

$$213 \quad (x^2 + y^2)^{\frac{1}{2}} - 10.5 \tan^{-1} \left(\frac{y}{x} \right) = 21 \quad (7)$$

214 From the x, y coordinates obtained from Equation 7, the CPC was designed in 3D (Figure 9a)
215 and finally its 3D printing (Figure 9b).

216 **Figure 9.** Compound Parabolic Cylinder Collector. a) 3D SketchUp® model. b) 3D printed PC.

217

218 3.3. Solar photodegradation of pollutants.

219 In the second stage, using the best conditions in SF degradation (air presence and high flow),
220 degradation tests of the 3 pollutants chosen for this research (SF, 2.4 - D and SAM) were carried out
221 using 1 g L⁻¹ of TiO₂ under solar radiation using the designed collectors until achieving 122.77 kJ m⁻² of
222 accumulated energy in the system. Figure 10 shows the pollutants degradations obtained with the use of
223 the 4 types of radiation collectors.

224

225

226 **Figure 10.** Degradations achieved using the 4 radiation collectors for the 3 pollutants.

227

228 First, it can be observed that the use of solar radiation collectors improves in significant percentages the
229 chosen molecules photodegradation for this study, since the percentages of degradation increase between
230 1.5 and 12 times the remotes obtained without them.

231 For SF, although a degradation of 90.82% was achieved with the CP (Figure 10), its enhancing effect was
232 only 1.89 times since its initial removal with a lamp was 48% under better treatment conditions. On the
233 other hand, when analyzing the behavior of 2,4-D, is why it can be observed that although the effect of
234 the use of collectors improved its degradation between 1.27 and 2.89 times (greater than for SF, Figure
235 10) only achieved a contaminant degradation of 33.66%, this is due to the high stability and low
236 degradability of the molecule (Álvarez 2007). Finally, in the case of SAM (molecule which the greatest
237 effect of the use of collector was obtained in its photodegradation), removals were achieved between
238 10.91 - 12 times those achieved with lamps without radiation collector (Figure 10), which may be due to
239 because this pollutant absorbs radiation in the visible spectrum, which favors its denaturation (Granda-
240 Ramírez 2017).

241 From the point of view designed collectors comparison, it is observed those although the best geometry
242 was the parabolic (Figure 10), in general there are not significant differences between their performances,
243 since for each of the molecules studied a similar photodegradation percentages was achieved,
244 demonstrating the importance of correctly designing the radiation collector in photocatalytic applications.

245

246 To determine the impact of the use of solar collectors, the Collector Impact Ratio Factor (CIRF) was
247 determined by the relationship between the degradation obtained with each collector by the degradation
248 obtained with the system without a collector. A ratio higher than 1 indicates an improvement in the
249 degradation system, less than 1 a decline in degradation, and a ratio equal to 1 indicates that the collector
250 does not affect degradation either positively or negatively (Figure 11).

251

252 **Figure 11.** Collector Impact Ratio Factor (CIRF) to SF, 2,4-D and SAM photodegradation using solar
253 radiation.

254

255 Figure 11 shows that in general, the use of collectors improved the photocatalytic system used
256 performance (Blanco 2004 and Doménech 2004), observing that the parabolic collector (PC) is the one
257 that most increases the degradation of the 3 pollutants, therefore This geometry type is one of the most

258 widely used generally in solar applications (Maldonado 2015 and Olleros 2013). Analyzing the behavior
259 of each pollutant in the designed collectors, it's can see that there was the greatest impact for SAM, in
260 which the degradation increase was around 11 to 12 times, this can be explained by the fact that SAM is
261 sensitive to visible light and solar radiation have about 45% of this kind of radiation (Hincapié-Mejía
262 2020). On the other hand, it can be observed that the molecule that has a lower value of the CIRF was the
263 SF, since the degradation improved between 50 and 89%, going from a degradation of 48% without
264 collector to 90.82% with the CP (Figure 10). Finally, although the CIRF value for 2,4-D is higher than
265 that of SF, its removal levels are not so high, since around of 33.66% were achieved in the best case.
266 (Figures 10 and 11), thus showing the great stability that this molecule has. Finally, it can be said that
267 although the performance of the CP was the best of the 4 collectors designed, actually the difference in
268 their performance is similar in each of the pollutants studied (Figures 10 and 11).

269

270 **4. Conclusions**

271 First, Dark adsorption test determined that there is a contaminant affinity with the photocatalyst, thus
272 facilitating its degradation at the time of irradiating the system. Second, aeration supplied to the
273 degradation system allows greater degradation of pollutants due to the formation of reactive oxygen
274 species (ROS) which potentiates degradation; also, It was possible to determine that the high flow favors
275 the pollutants degradation since this, having a speed 3 times higher than the low flow, lets a radiation
276 longer exposure time of the solution to be treated, thus achieving a better contaminants remotion. Respect
277 to the using solar radiation intensified with collectors improves the pollutants degradation due to its
278 adequate design, especially with the most resistant molecules under the conditions evaluated, since
279 improvements were obtained in the degradation of pollutants up to 12 times that achieved without
280 radiation collectors. Finally, although the collector with the best performance was the parabolic (PC), no
281 significant differences were found in the magnification of the use of solar radiation by the collectors
282 designed for the photochemical processes studied in this research.

283

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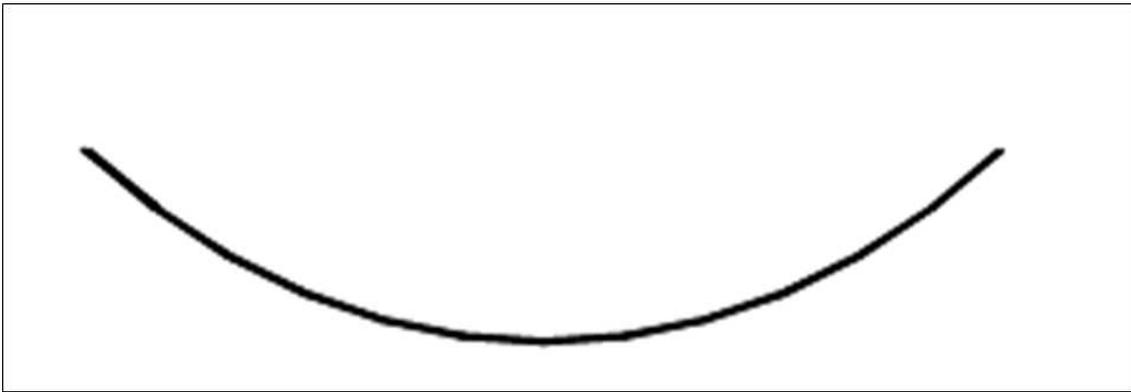
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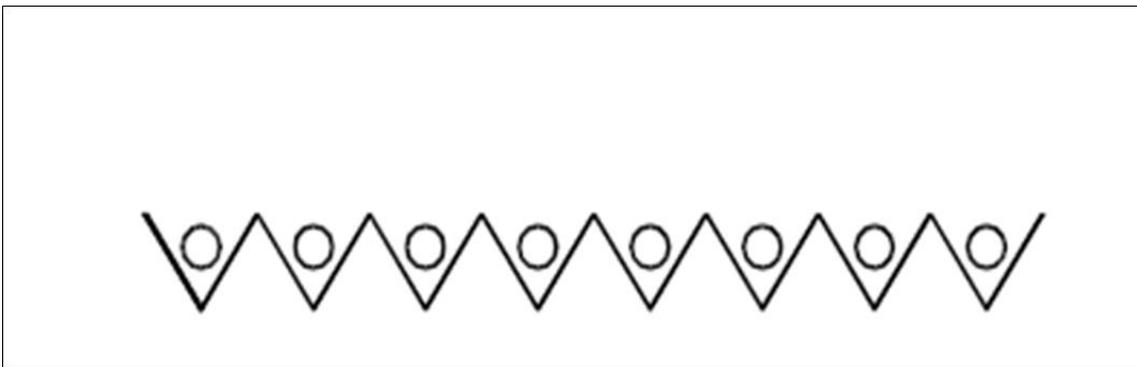
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340 **Figure 1.** Parabolic Collector (PC).

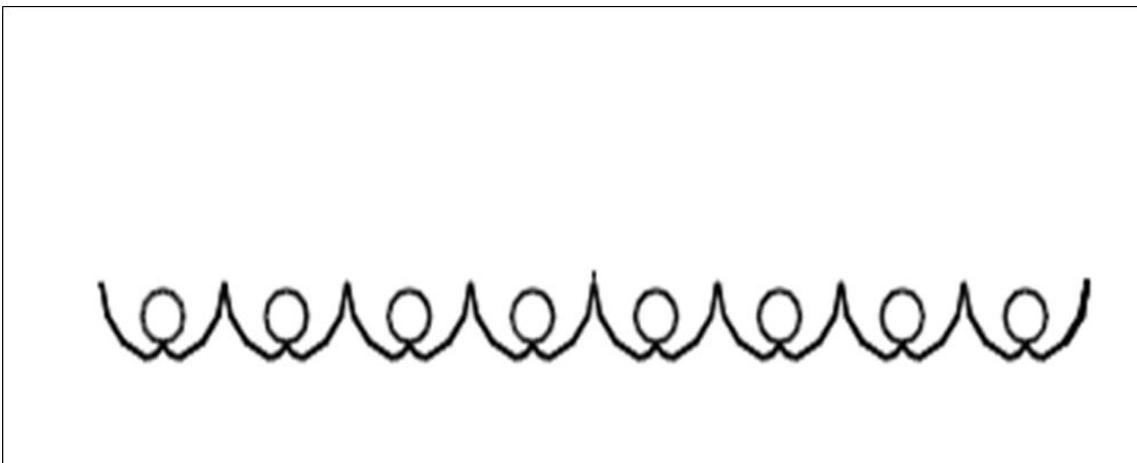
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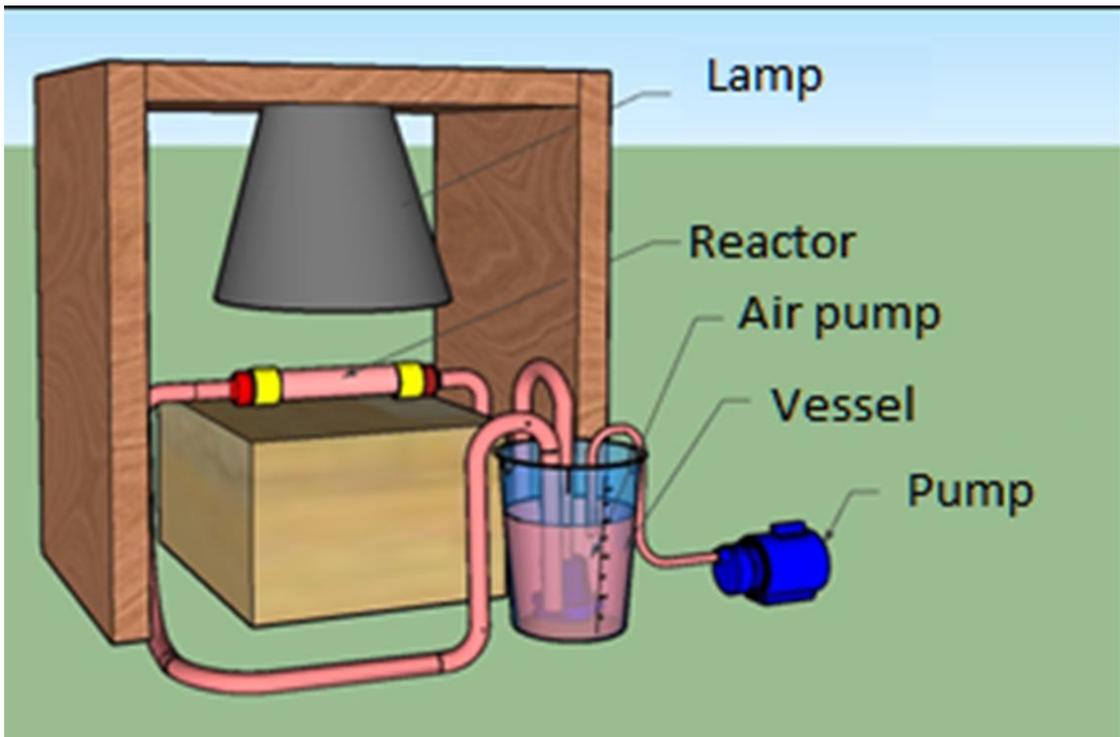
343 **Figure 2.** V Collector (VC).

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346 **Figure 3.** Compound Parabolic Cylinder Collector (CPC).



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348 **Figure 4.** First stage's reaction system.

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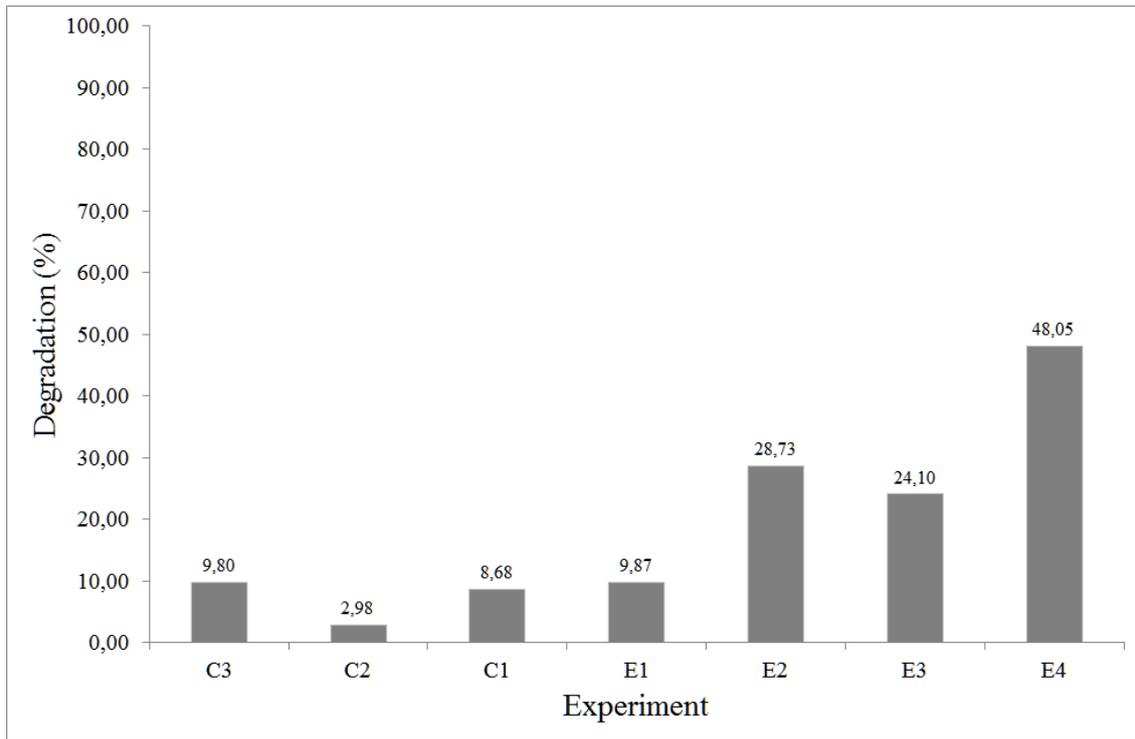
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361 **Figure 5.** First stage results to determine the best conditions for SF degradation.

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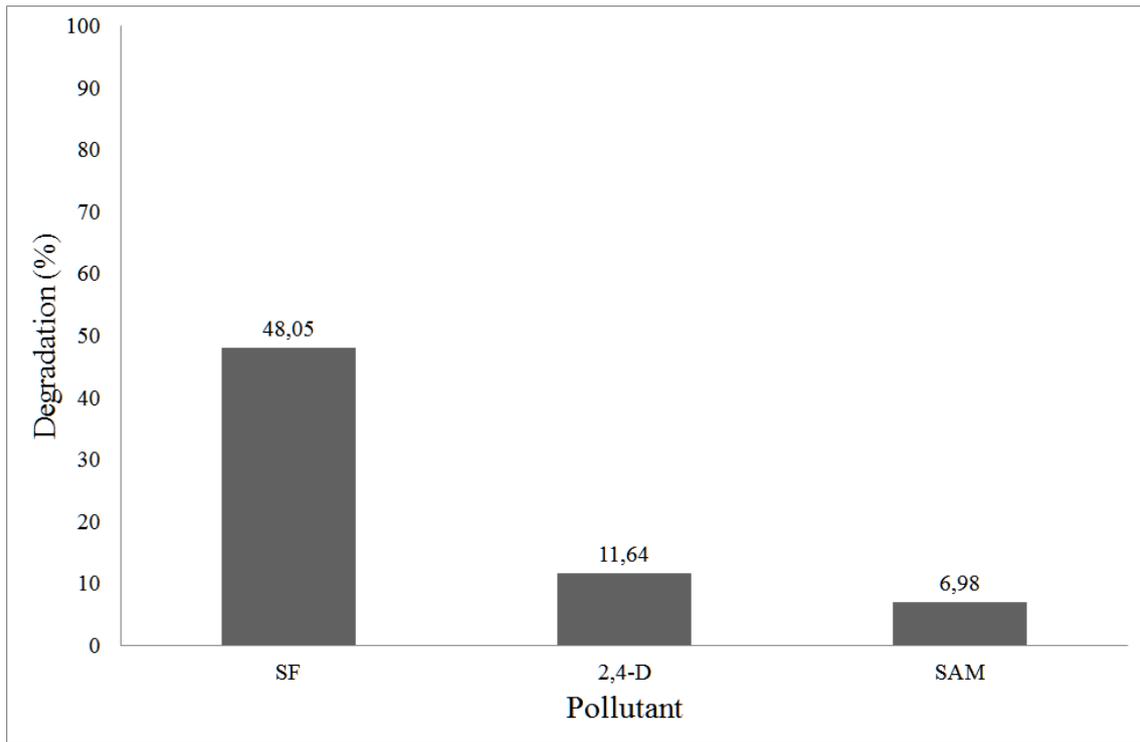
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374 **Figure 6.** CEC degradation in best conditions using a UV lamp without collector.

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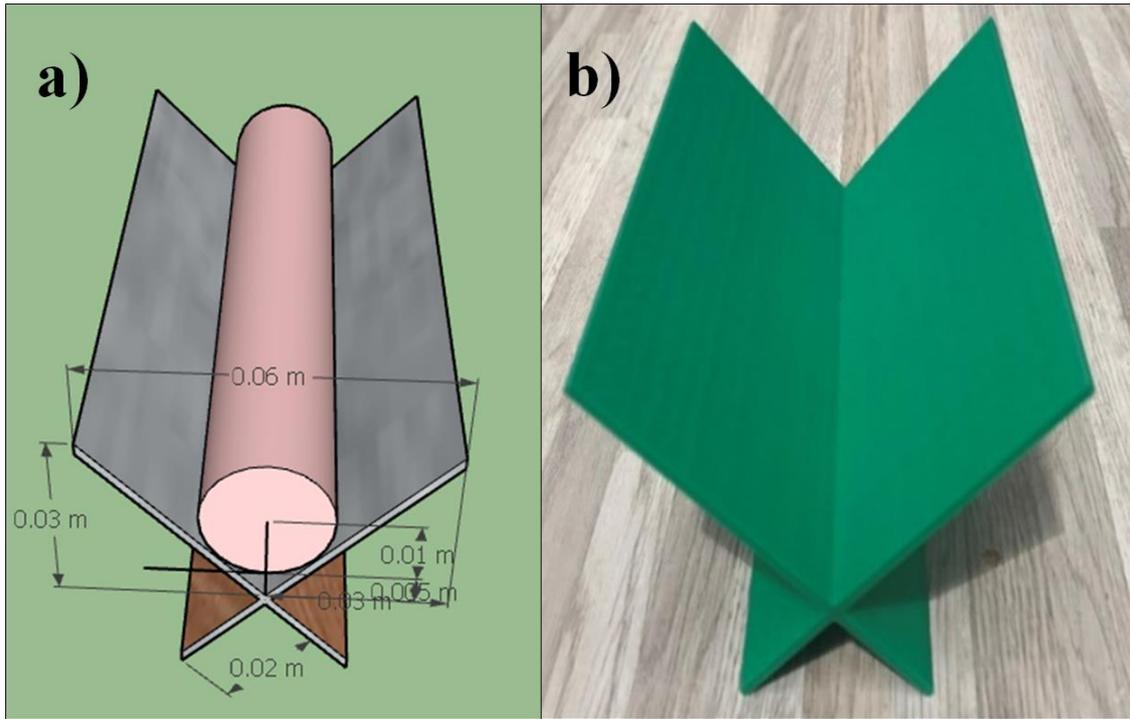
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387 **Figure 7.** V Collector. a) 3D SketchUp® model. b) 3D printed VC.

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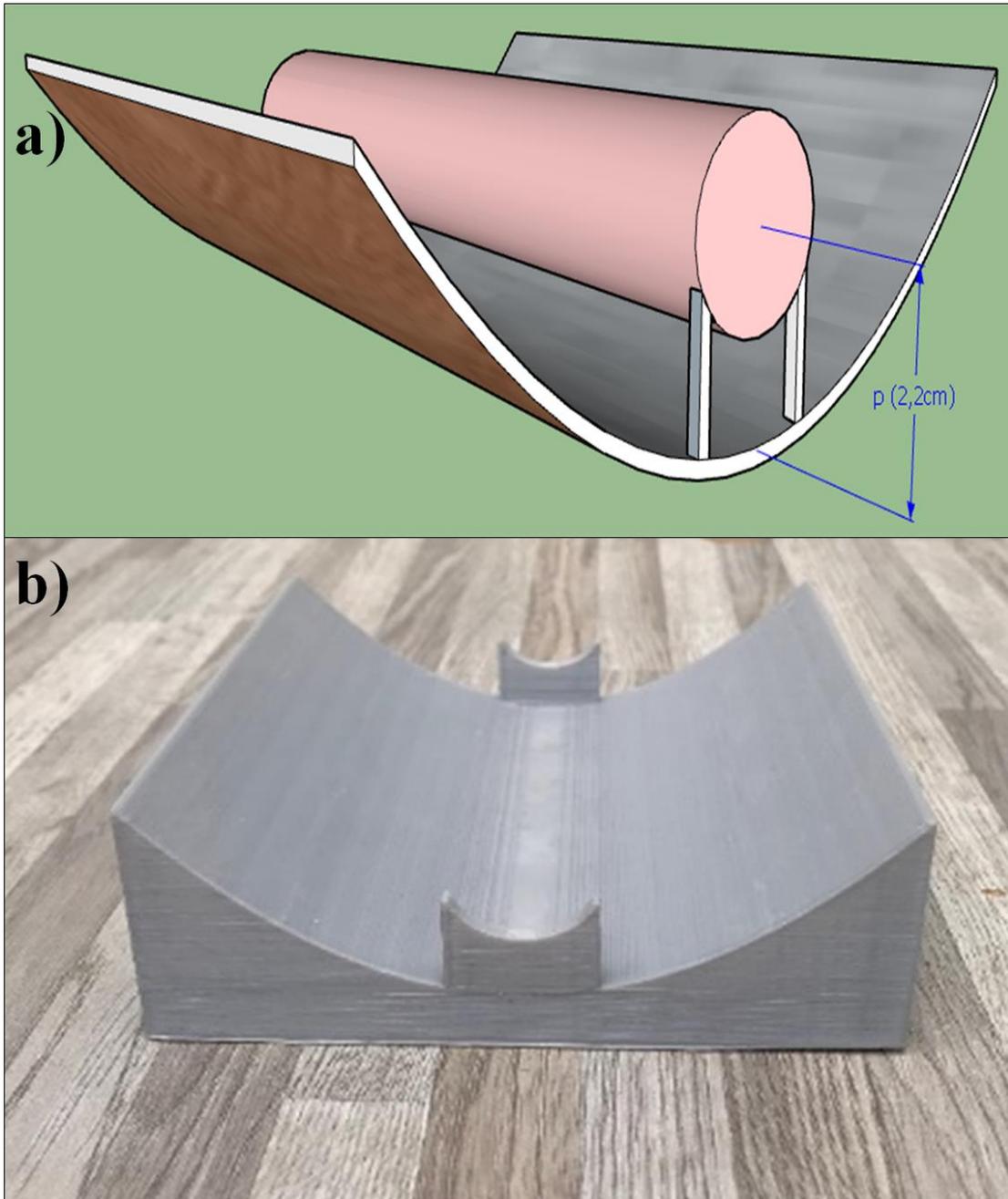
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401 **Figure 8.** Parabolic Collector. a) 3D SketchUp® model. b) 3D printed PC.

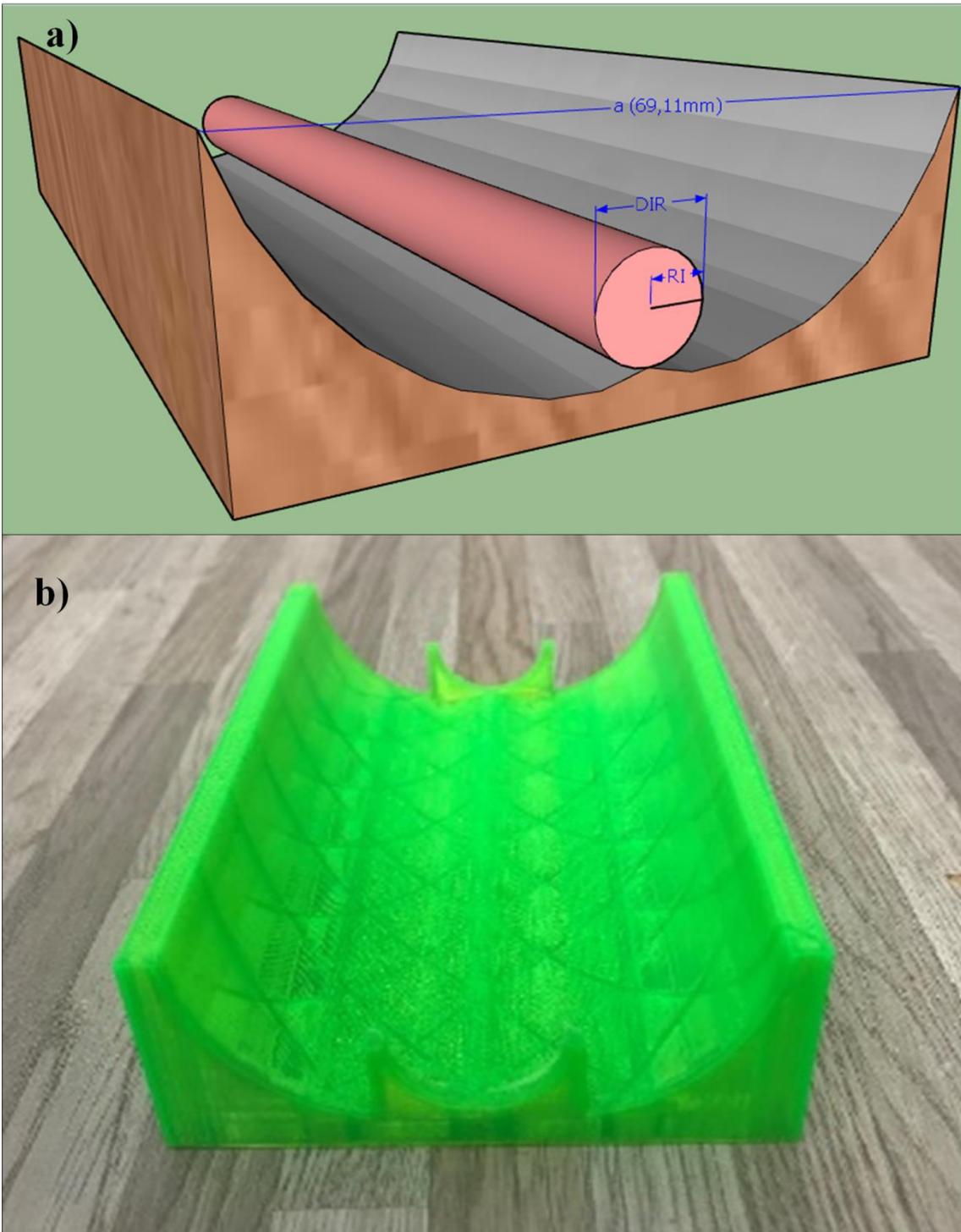
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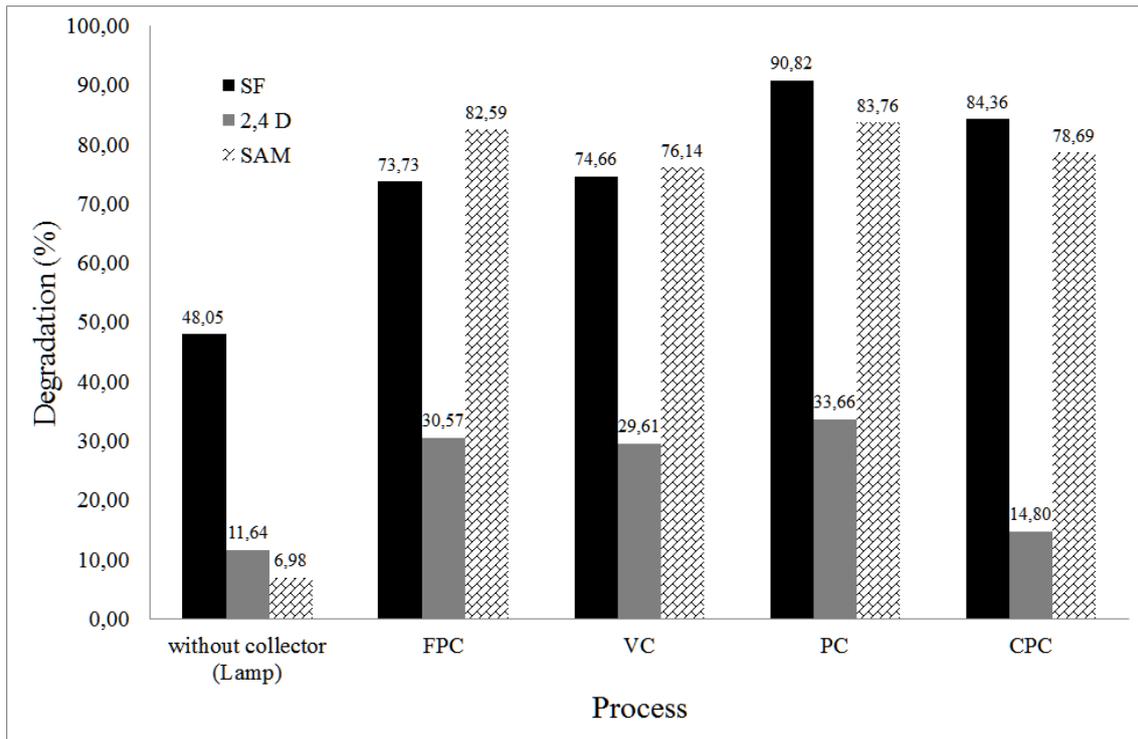


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408 **Figure 9.** Compound Parabolic Cylinder Collector. a) 3D SketchUp® model. b) 3D printed PC.

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412 **Figure 10.** Degradations achieved using the 4 radiation collectors for the 3 pollutants.

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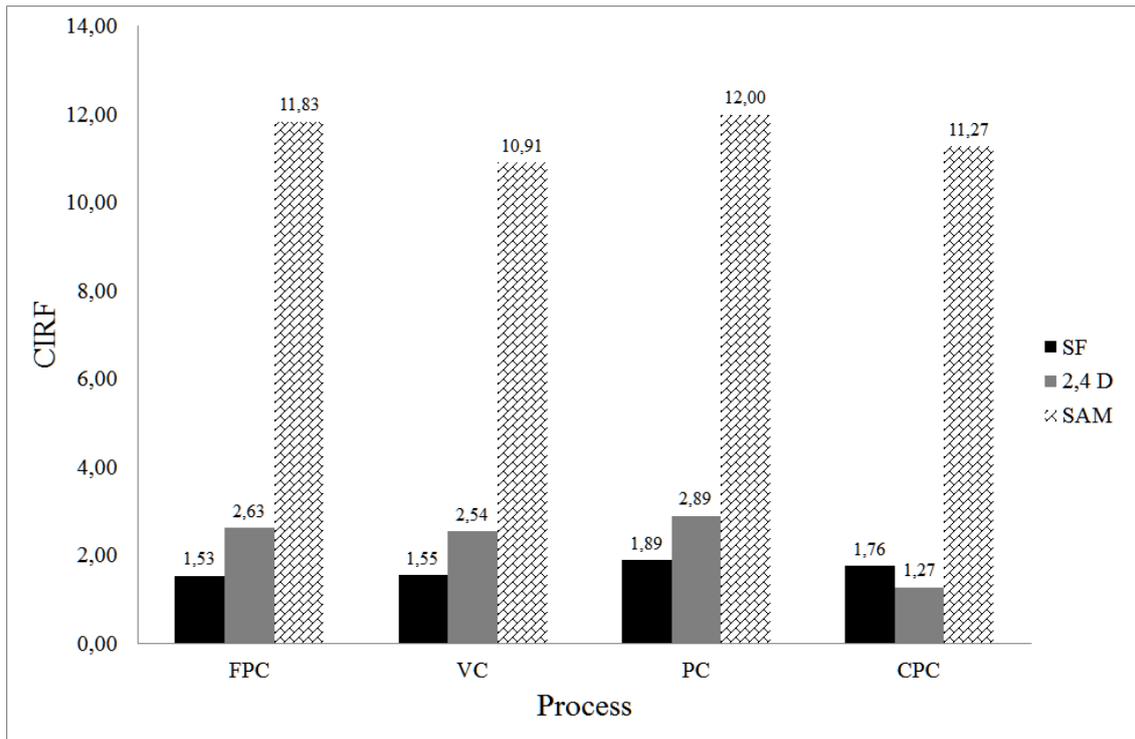
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425 **Figure 11.** Collector Impact Ratio Factor (CIRF) to SF, 2,4-D and SAM photodegradation using solar
 426 radiation.