Title: Fresh-cut wastewater reclamation: techno-economical assessment of solar driven processes at pilot plant scale

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ABSTRACT

Up-scaling of solar processes for water purification is a key challenge for their implementation at industrial scale on the agro-food sector. Benefits of using flow-tubular reactors provided with Compound Parabolic Collector mirrors has been previously demonstrated, nevertheless some techno-economic aspects still being unknown.

This study shows a comparative analysis of the treatment efficiency of H_2O_2/solar and Fe^{3+}-EDDHA/solar and Fe^{3+}-EDDHA/H_2O_2/solar as novel processes for treating synthetic fresh-cut wastewater (SFCWW) containing 100 NTU of turbidity. The highest treatment capability was obtained with Fe^{3+}-EDDHA/H_2O_2/solar (2.5/20 mg/L-Fe^{3+}-EDDHA/H_2O_2), attaining the fastest microbial inactivation kinetics (>5-log of *E. coli* O157:H7 and *Salmonella enteritidis*) and OMCs degradation (36% of 5 microcontaminants) in 60 and 120 min, respectively.

Treated SFCWW by Fe^{3+}-EDDHA/H_2O_2/solar process fits microbiological quality established in water reuse guidelines for irrigation, no bacterial reactivation after 24h post-treatment, no significant ecotoxicity and treatment cost was estimated as 1.10 (only disinfection) and 2.10 €/m^3 (simultaneous disinfection and decontamination).

Keywords: Agro-food sector, Foodborne pathogens, Compound Parabolic Collector, Near-neutral pH, Iron-chelate.

1. Introduction

The fresh-cut produce industry has suffered a fast growth in the last years, expecting to continue due to the increase of the global population and consumer's shifts toward ready-to-eat fresh food (like leafy salads) that stand out in healthy diets [1]. This industry is one of the major water consumers of the agro-food sector due to the high water volumes (up to 40 m^3/ton of raw product) required during processing stages (prewashing, disinfection and rinsing of the vegetables). The water quality for this sector must accomplish the microbial and chemical quality standards of the potable water (Council Directive 98/83/EC (1998)) [2] explained by the fact of the consumption of raw vegetables, a potential risk for consumers if a contamination event occurs during the processing steps. In fact, several foodborne outbreaks mainly caused by the faecal pathogens *E. coli* O157:H7, *Salmonella* spp, *Listeria monocitogenes*, and some viruses have been reported during last several years associated to this industry [3]. In this regard, is important to note that most of the infections outbreaks reported are caused by *E. coli* O157:H7 and *Salmonella spp* which are considered the main food-borne pathogens during production chain of fresh-products. On the other hand, along the processing vegetables, the wash-water generated also represents an important source of organic microcontaminants (OMCs), mainly by pesticides employed during the crops cultivation which are continuously released in the processing-water and detected in the range of nano to microgram per liter. This highlights the importance of controlling the OMCs accumulation during the processing stage of vegetables and its discharge as a preventive action to reduce further potential health and environmental impacts [4].

Chlorination is widely used as water disinfection treatment in this industry, but the high amount of organic matter in the fresh-cut washing water and the usually hyper-chlorination practice leads to the generation of high amounts of unhealthy disinfection by-products (DBPs) and to the forbiddance of the chlorination practice in some European countries [5]. Besides, this treatment did not remove OMCs from water. Therefore, the implementation of a sustainable water management system to efficiently control water-pathogens, OMCs and the formation of disinfection-by-products (DBPs), providing a regenerated wastewater with a quality enough to be reused, can lead to an improvement of the water foot-print of this industry [6]. Accordingly, the search and evaluation of alternative water treatments has grown in last years including electrolyzed water, chlorine dioxide, organic acids, UV-C, quaternary ammonium compounds (QACs), essential oils, ozone, and cold plasma among others, although no great efficiencies have been reported [7]. Other processes like the Advanced Oxidation Processes (AOPs) have shown high disinfection capability in a wide range of water matrices including fresh-cut

washing water, like UV-C/H_2O_2, UV-C/PAA [8], UV-C/ozone [9] or UV-C/TiO_2 [10] due to the formation of powerful oxidants such as hydroxyl radicals ($^{\bullet}OH$). However, all of them use UV-C lamps as a photons source which generates high process costs and represents one of the main drawbacks of their implementation. Moreover, there is still a lack of information about their capability to simultaneously disinfect and degrade OMCs in clear and turbid waters.

To this respect, the use of solar radiation as source of photons, the so-called solar-driven AOPs and solar photochemical processes, have demonstrated a high decontamination and disinfection efficiency including a wide range of water matrices [11]. To date, the photo-Fenton process is one of the most investigated solar AOP. It uses a combination of iron salt, H_2O_2 and solar radiation to generate $^{\bullet}OH$ [12]. Recently, to increase the efficiency of this process at near neutral pH, the use of complexing agents that allow to keep the iron in solution have been encouraged, including both synthetic [13] and natural chelating agents [14]. The synthetic chelating agents based on aminopolicarboxylic acids like EDTA (Ethylendiaminetetracetic acid) and EDDS (Ethylendiamine-N‘,N‘-disuccinic acid) have shown a good performance based on their high stability [15]. More recently, the use of EDDHA (Ethylenediamine-N,N'-bis 2-hydroxyphenylacetic acid) as Fe-chelate agent for disinfection of synthetic FCWW at near-neutral pH under natural sunlight has been reported at laboratory scale [16]. Another promising solar process performed at near-neutral pH for water disinfection is the combination of H_2O_2 with natural sunlight (H_2O_2/solar) due to the very well reported accelerated effect on the inactivation kinetics of different microorganisms, requiring the addition of low concentration of reagent (range 0.1-0.3 mM) in different water matrices, including urban and fresh-cut wastewater [17-19].

In addition, the development of solar Compound Parabolic Collector (CPC) reactors has demonstrated to be the most promising technological solutions for application of solar water treatments at industrial scale [20,11]. Among its main advantages highlight its high efficiency on the use of solar photons, with the respective acceleration of the kinetics reactions, and the modular design that permits increase the volume of water intended to be treated and its easily installation and adaptation as decentralized system.

The aim of this study is, therefore, to assess for the first time the up-scaling (to dozens of litres) of several solar processes for the simultaneous disinfection and decontamination of synthetic fresh-cut wastewater (SFCWW) using solar reactors provided with CPC mirrors. In particular, three solar processes were investigated; H_2O_2/solar and two novel photo-Fenton processes based on the effect of a chelating agent: Fe^{3+}-EDDHA/solar and Fe^{3+}-EDDHA/H_2O_2/solar at near neutral pH. A techno-economic assessment of the feasibility of these solar processes has

been performed based on their efficiency to simultaneously inactivate two pathogens (*E. coli* O157:H7 and *Salmonella enteritidis*) and to remove 5 OMCs. As complementary to the overall efficiency of the solar processes, a regrowth analysis and an acute toxicity assessment were also performed, providing a realist insights of the feasibility of the proposed solar processes as alternative treatment to chlorine in FCWW. This study demonstrates the potential possibility to employ an iron chelate already added by farmers in water-scarce regions to regenerate FCWW contributing at the same time to remediate two environmental problems in arid and semiarid regions (such as Mediterranean countries): water scarcity and iron chlorosis disease.

2. Materials and methods

2.1 Water matrix

The water matrix used was synthetic fresh-cut wastewater (SFCWW), which permits a comparison between the different processes avoiding the effect of the composition variation of the industrial washing water. This is a lab-made synthetic water daily prepared following a recipe previously developed and based on literature data and the analysis of real samples from a local fresh-cut industry. This recipe is fully described elsewhere [19]. Its main physicochemical characteristics are: 100.1±0.4 NTU of turbidity, 6.25±0.06 water pH, 1209.6±14.8 μS/cm of conductivity and 45 mg/L of dissolved organic carbon (DOC) (corresponding to a 163 mgO_2/L of Chemical Oxygen Demand). The DOC value correspond to the sum of 25.4±0.4 mg/L from the organic matter of water recipe and 20 mg/L from the organic solvent used for OMCs preparation). Moreover, the addition of the iron chelate increases the water DOC content: ≤ 2, 9 and 18 mg/L for 0.5, 2.5 and 5 mg/L of iron as Fe^{3+}-EDDHA (1:1), respectively. The absorbance spectrum of the SFCWW with and without the presence of the iron chelate was measured in the range 200-800 nm by using a UV-spectrophotometer (Thermo Scientific Evolution 220).

2.2 Bacteria quantification

E. coli O157:H7 (CECT 4972) and *Salmonella subsp. enteritidis* (CECT 4155) from the Spanish Culture Collection (CECT) were used as models of microbial contamination. Quantification and enumeration methods were done as described elsewhere [19]. Briefly, liquid bacterial cultures containing a bacterial concentration of 10^9 CFU/mL were prepared using Nutrient-Broth I (5 g/L of NaCl (Sigma Aldrich), 5 mg/L beef extract and 10 g/L of peptone (Panreac, Spain)) for *E. coli* O157:H7 and Tryptone Soya Broth (OXOID) for *S. enteritidis*.

Bacterial concentration was enumerated by the standard plate counting technique using serial dilutions (10 fold) in phosphate buffer saline (PBS). For that, sample volumes of 50 µL and 500 µL (detection limit (DL) of 2 CFU/mL) were spread on ChromoCult® Coliform Agar (Merck KGaA, Darmstadt, Germany) and Salmonella Shigella Agar (Scharlau®, Spain) and incubated at 37ºC during 24h and 48h for *E. coli* O157:H7 and *S. enteritidis*, respectively.

Besides, membrane filtration method was used to assess the regrowth of bacteria after 24 h of storage in the dark. In this case, 100 mL of sample was filtered using a Microfil®filtration system (Millipore, USA) and cellulose nitrate filters (0.45 µm, Sartorius Stedim, Spain) using similar culture media procedure as described previously, reaching a DL of 1 CFU/100mL to fit with the limit established in guidelines for wastewater reuse [21-22].

2.3 Reagents and analytical determinations

The commercial micronutrient Sequestrene 138 Fe G100 (source of iron containing 6.2% soluble iron content as Fe^{3+}-EDDHA, Syngenta, Spain) [16] and hydrogen peroxide (35% w/v, Merck, Germany), were used as received from the manufacturer. Fe^{3+}-EDDHA was used as source of iron due to this iron chelate is the synthetic iron fertilizer most employed in agriculture to prevent and remediate iron chlorosis disease [23].

Dissolved iron and H_2O_2 concentration were measured by spectrophotometric methods following the procedure described elsewhere [24]. To measure the dissolve organic content (DOC), the water samples were filtered using a 0.2 µm nylon syringe-driven filter (Millex) and measured by a Total Organic Carbon analyzer (Model 5050, Shimadzu, Japon).

Quantification of OMCs (atrazine, azoxystrobin, terbutryn, procymidone and buprofezin) was done by liquid chromatography using an Agilent 1260 (Palo Alto, CA, USA) with a diode array detector (UV-DAD) and a C-18 column (XDB-C18, 1.8 µm, 4.6x50 mm). The eluents were acetonitrile (ACN) (HPLC grade, Panreac, Spain) and acid ultrapure water (25 mM formic acid) with a flow rate of 1 mL/min and an injection volume of 100 µL. The working method consists on: 0.5 min of isocratic condition (90% H_2O:10% ACN), followed by 5.5 min of a linear gradient to 100% ACN, 100 % ACN during 1.5 min and returning to the initial conditions in 1 min. Before injection, 4.5 mL of water sample was filtered using a 0.2 µm syringe-driven filter of nylon (Millex), and after that 0.5 mL of ACN was passed through the filter to remove any possibly adsorbed compounds. The detection wavelength and limit of quantification (LOQ) of the different OMCs were 250 nm for buprofezin (LOQ: 5 µg/L), 230 nm for atrazine (LOQ: 2 µg/L) and terbutryn (LOQ: 1.9 µg/L) and 214 nm for azoxystrobin (LOQ: 20 µg/L) and procymidone (LOQ: 3.6 µg/L). These pesticides were selected as OMCs targets for this study

according to their wide use during vegetables growing (cultivation) and their inclusion (atrazine and terbutryn) as priority substances (PS) in the latest European directives.

2.4 Solar experiments

Solar experiments were performed at Plataforma Solar de Almeria (South-East of Spain) during May-July 2018. All the tests were performed in a solar Compound Parabolic Collector (CPC) photoreactor placed on an anodized-aluminium platform titled at 37°. This CPC reactor has been widely described elsewhere [25]. Briefly, it is formed by two CPC mirror modules made by highly reflective anodized aluminium (MiroSun, Alanod, Germany) with a concentration factor of 1. Each module has 10 borosilicate-glass tubes (1500 mm x 50 mm and 2.5 mm thick) with a total irradiated surface of 4.5 m^2 and 75 % of total water volume irradiated (45 L out of 60 L). The water is recirculated through the tubes by a centrifugal pump (150 W, Mod.NH-200 PS PanWorld, USA) with a flow rate of 30 L/min. The pH and water temperature are monitored throughout the experiments by sensors placed in the dark piped-system. No significant pH variations (6.25±0.3) were observed along the treatment time for any of the conditions tested. Reagents, OMCs and microbial suspensions were directly and simultaneously diluted in the reactor to obtain the desired initial concentration: 100 µg/L and 10^6 CFU/mL for each OMC and pathogen, respectively. Reagents concentrations from 2.5 to 40 mg/L of H_2O_2 and 0.5 to 5 mg/L of Fe^{3+}-EDDHA were selected according to previous studies and considering the range of the iron micronutrient concentration usually employ in intensive agriculture [16]. The water was homogenised during 15 minutes in the dark. After that, the first sample was taken out and immediately the reactor was exposed to sunlight. All solar experiments started between 10:15-10:45 a.m. local time and lasted 5 hours under full sunshine. The water samples were taken out at regular intervals for OMCs and bacteria analysis. To evaluate the mere effect of the solar processes studied in the selected targets, the residual H_2O_2 concentration was eliminated in the samples by the addition of a bovine liver catalase solution (0.1 g/L) at ratio catalase:sample of 1:50 avoiding therefore any oxidative post-treatment effect during the time of samples analytical procedure [19]. The averaged day-profile of water temperature and solar UVA radiation registered during all solar treatments is shown in Figure SI1 (Supplementary information). The water temperature ranged from 25±4 ºC to 41±6 ºC for all the experiments, discarding therefore thermal inactivation of bacteria [26]. Solar UV-irradiance (λ: 280-400 nm) was monitored by a pyranometer (Kipp&Zonen, CUV-5, Netherlands) which provided irradiance data in W/m^2. Maximum and minimum solar UV irradiances were 26±3 and 49±3 W/m^2, respectively.

The microbial inactivation and OMCs degradation were plotted as the average values of two replicates (standard deviation as error bar) against the accumulative UV energy during exposure time per unit of treated water volume (Q_{UV}; kJ/L), calculated according to a previous works [25].

2.5 Inactivation kinetic analysis

Kinetic constants were calculated considering the Q_{UV} parameter instead of the treatment time. The kinetic rates of bacterial inactivation were obtained by several mathematical models described elsewhere [27] and according to the higher R^2 value fitting the experimental data:

1) A log-linear decay according to the Chick´s law (Eq. 1).

2) A 'shoulder phase' characterized by a constant bacteria concentration or a very smooth decay followed by a log-linear decay, attributed to the accumulation of oxidative damages ending in the loss of cells viability (Eq. 2).

3) A double log-linear kinetic characterized by a fast inactivation in the first stage (k_1) followed by a slow second inactivation stage (k_2) (Eq. 3).

4) A log-linear decay followed by a 'tail' (Q_{UVres}) (Eq. 4). The 'tail' represents the bacterial population that remains at the end of the experiment due to the presence of a resistant population. In our case, the tail observed may be due to the presence of dark zones in the reactor (25 % of the total volume) which enable bacteria to repair partially their oxidative damage.

5) A 'shoulder phase' followed by a log-linear decay and a 'tail' (Eq. 5).

$$Log\left(\frac{N}{N_0}\right) = -k \cdot Q_{UV} \qquad \text{Eq.1}$$

$$Log\left(\frac{N}{N_0}\right) = -k \cdot Q_{UV} \quad \begin{Bmatrix} 0 \;\; ; Q_{UVt} \geq Q_{UV} \\ -k \cdot Q_{UV} \;\; ; \;\; Q_{UVt} < Q_{UV} \end{Bmatrix} \qquad \text{Eq.2}$$

$$Log\left(\frac{N}{N_0}\right) = -k_1 \cdot Q_{UV}\,; Q_{UV} = [0, Q_{UV1}]; \quad Log\left(\frac{N}{N_0}\right) = -k_2 \cdot Q_{UV}\,; Q_{UV} = [Q_{UV1}, Q_{UV2}] \qquad \text{Eq. 3}$$

$$Log\left(\frac{N}{N_0}\right) = -k \cdot Q_{UV} \quad \begin{Bmatrix} -k \cdot Q_{UV} \;\; ; \;\; Q_{UVt} \leq Q_{UV} \\ 0 \;\; ; \;\; Q_{UVt} \leq Q_{UVres} \end{Bmatrix} \qquad \text{Eq.4}$$

$$Log\left(\frac{N}{N_0}\right) = -k \cdot Q_{UV} \quad \left\{ \begin{array}{l} 0 \;\; ; \;\; Q_{UVt} \geq Q_{UV} \\ -k \cdot Q_{UV} \;\; ; \;\; Q_{UVres} < Q_{UVt} < Q_{UV} \\ 0 \;\; ; \;\; Q_{UVt} \leq Q_{UVres} \end{array} \right\} \qquad \text{Eq.5}$$

N/N_0 is the bacteria concentration along the solar treatment, k is the disinfection kinetic constant and Q_{UVres} is the energy value with a residual bacterial population.

2.6 Toxicity assessment

Aliivibrio fischeri was used as a test organism for the assessment of toxicity, this is a widely used organism for initial screening in environmental samples with unknown ecotoxicological characteristics [28]. Samples from treated SFCWW were collected at the end of each solar processes and directly used for the analysis of toxicity, no catalase was added in this case in order to determine the possible toxic effect of residual reagents in the treated water. The assessment of acute toxicity was carried out by monitoring changes in the bioluminescence of *A. fischeri* after 30 min of contact with water samples. Prior to the test, water pH of each sample was adjusted to 6~7.5, filtered with 0.2 μm syringe-driven filters (Millex®, Millipore) and salinity adjusted to 2 % (w/v). Samples were tested in triplicate. The 30-min luminescence inhibition test was performed according to standardized protocols [29]. The bioluminescence of *A. fischeri* was measured using the BioFix® Lumi-10 luminometer (Macherey-Nagel GmbH & Co. KG, Duren, Germany). Toxicity results were expressed as bioluminescence inhibition percentage (BI %) and samples were considered toxic when this value was above 50 % [30].

3. Results and discussion

3.1 Bacterial inactivation by solar processes

3.1.1 Solar only disinfection and H_2O_2/solar process

Prior to any solar test, the potential effect of OMCs over the bacterial viability was assessed in the dark for 300 min of treatment time, results revealed no decrease on the initial bacterial concentration, discarding therefore any effect of OMCs over the kinetics of the microorganisms.

The inactivation kinetic profiles of *E. coli* O157:H7 and *S. enteritidis* at pilot plant scale by H_2O_2/solar process at several concentrations (2.5, 5, 10, 20 and 40 mg/L) are shown in Figure 1a and 1b, respectively. Also, the mere effect of solar radiation (solar only disinfection) on the inactivation of both pathogens was investigated. The corresponding inactivation kinetic

constants are summarized in Table 1. Data of H_2O_2 concentration measured are shown in Table 2 and Figure SI2 (Supplementary material). The residual concentration of H_2O_2 at the end of each solar process ranged from 63 to 77 % of the initially added in all cases.

The solar only disinfection process leads a similar kinetic behavior in both pathogens characterized by an initial log-linear decay followed by a residual concentration of bacteria, not attaining a complete removal (DL: 2 CFU/mL) after 300 min of solar exposure. This inactivation kinetic profile has been reported previously in other studies at pilot plant scale [31]. The residual population remaining in the systems is attributed to the low-oxidative capability of this treatment and the interrupted delivered solar UV radiation due to the re-circulation of the water through the dark and illuminated areas of the solar CPC reactor during the solar exposure. Both effects may permit the activation of the self-defense mechanism of bacteria to repair the oxidative damages generated inside cells during the solar exposure, keeping therefore a residual population in the sample resistant to be inactivated by solar radiation [31].

This residual effect can be avoided by applying a more oxidative solar process, such as the H_2O_2/solar process, which results clearly show an enhancement of the bacterial inactivation profiles (Figure 1a,b) and kinetic constants (Table 1) compared with solar only disinfection process. In this case, DL was achieved for both pathogens with all the H_2O_2 concentrations tested. In general, the higher the H_2O_2 concentration, the higher the inactivation kinetic. This improvement was marked at H_2O_2 concentration values of 20 mg/L (Table 1) for both pathogens, especially in *S. enteritidis* where the inactivation kinetic profile changed from double log-linear decay to a log-linear decay. Nevertheless, *S. enteritidis* showed no-significant differences on kinetic rates by increasing the oxidant concentration from 20 to 40 mg/L. This behavior is in line with previous lab-scale results that suggested that the inactivation of *S. enteritidis* by H_2O_2/solar process is not-chemically limited at the range of reagents concentration tested [19]. The negligible oxidative effect of the H_2O_2 concentrations used in this study over bacteria viability has been previously reported [19], therefore, the bacterial inactivation shown in Figure 1 can be only attributed to the effects of the solar processes investigated.

Therefore, considering the overall data obtained from this solar process and the concentrations of H_2O_2 tested, the best inactivation rate was obtained with 20 mg/L of reagent for both bacteria. At this concentration, *S. enteritidis* showed a higher resistance to be inactivated (DL reached in 60 min of treatment time and 11.9 kJ/L of Q_{UV}) than *E. coli* O157:H7 (DL reached in 45 min of treatment time and 8.7 kJ/L of Q_{UV}). This difference can be attributed to their

different response against the oxidative stress generated by the solar process. It is believed that the bacterial inactivation mechanism by H_2O_2/solar process is based on a synergistic effect between the photo-oxidative damages induced by solar radiation and internal photo-Fenton reactions between intracellular iron and the diffused H_2O_2 across the cell membrane [18]. Although this bactericidal mechanism is similar for different bacterial strains, the response to the oxidative stress generated may varied for each particular microorganism. In our specific case, the upregulation and a higher induction level of some genes and enzymes which act against the oxidative stress generated by an excess of H_2O_2, such as OxyR related genes and Superoxide Dismutase enzyme (Mn-SOD) in *Salmonella* strain, could explain the higher resistance of *S. enteritidis* to be inactivated by the H_2O_2/solar process compared with *E. coli* O157:H7 [19].

This results demonstrate the highly efficiency of H_2O_2/solar process for SFCWW disinfection, even considering the presence of high turbidity, requiring values of 12 kJ/L of solar energy to reach 5-log reduction value of both bacteria. In comparison with our previous works reporting SFCWW disinfection by H_2O_2/solar process at lab scale, 200 mL of volume and 10 mg/L of H_2O_2, where a Q_{UV} of 12.5 kJ/L was required to reach same disinfection results [16], it is also clear the high performance of this solar-driven process for the treatment of turbid wastewater at pilot plant scale using solar CPC reactors.

3.1.2 Fe^{3+}-EDDHA/solar process

Figure 2a,b shows the inactivation profile of *E. coli* O157:H7 and *S. enteritidis* by Fe^{3+}-EDDHA/solar process at three different concentrations: 0.5, 2.5 and 5 mg/L. The dissolved iron concentration was measured along the solar processes (Table 2, Figure SI2). At the end of solar tests, more than the 30% of the initially iron added was still detected in the sample, demonstrating the benefits of using this chelate to keep the iron dissolved in water.

The inactivation profile of both bacteria showed that the presence of the iron-chelate provoked the appearance of a shoulder phase and lower kinetic constants compared with the effect of solar only disinfection (Fig. 1a,b, Table 1), but also at the same time it is able to reach the DL avoiding the non-desired residual concentration of bacteria observed for the solar only disinfection process. These results and behavior agree with the reported in a previous study at laboratory scale for similar Fe^{3+}-EDDHA concentration [16]. The inactivation mechanism that explain this improvement has been previously proposed and attributed to alterations of the bacteria membrane permeability through chelation (by EDDHA or other subproducts) of the stabilizing cations (Mg^{2+} and Ca^{2+}) in the liposaccharides layers of the bacterial membrane

[16]. Thus, the membrane alteration increases the bacteria's susceptibility to be inactivated by solar radiation as observed in the elimination of the bacterial residual phase by this solar process.

The inactivation results showed an improvement for iron chelate concentration increased from 0.5 (no DL reached in any pathogen) to 2.5 mg/L; meanwhile, an opposite behavior occurred when the Fe^{3+}-EDDHA concentration raised from 2.5 to 5 mg/L appearing a detrimental effect on the inactivation of both pathogens (higher Q_{UV} need to reach the DL). The best inactivation results were therefore obtained with 2.5 mg/L of Fe^{3+}-EDDHA concentration. *S. enteritidis* (*k*: 0.229±0.029 L/kJ) showed also a higher resistance to be inactivated than *E. coli* O157:H7 (*k*: 0.417±0.026 L/kJ), requiring 30 min of treatment time and 6 kJ/L more of Q_{UV} to reach the DL.

The inactivation results have a clear dependency on the concentration of iron-chelated added. The lower efficiency showed by increasing the iron chelate concentration from 2.5 to 5 mg/L can be explained by an increase of the light scattering effect of the precipitated iron particles from the Fe^{3+}-EDDHA decomposition [32] that may act as a solar-screen protecting the bacteria from the solar UV-photons (a significant turbidity increase of ca. 5 NTU was observed at the end of the treatment for the highest iron concentration tested). In fact, the particle concentration (reagents not dissolved) is as key parameter in photocatalysis, especially for semiconductors such as titanium dioxide (TiO_2) and also for solar photo-Fenton and its behaviour has been very well described and modelled in tubular CPC photo-reactors, where it has been previously reported that an iron concentration higher than 5.5 mg/L (0.1 mM) can reduce the disinfection efficiency [27]. This detrimental effect is mainly a consequence of the particles screen effect that modify the homogeneous distribution of photons in the tubular reactor, a key feature of the treatment performance of CPC reactors.

3.1.3 Fe^{3+}-EDDHA/H_2O_2/solar process

The inactivation profiles of *E. coli* O157:H7 and *S. enteritidis* in SFCWW by the Fe^{3+}-EDDHA/H_2O_2/solar process are shown in Figure 3a and b, respectively. The inactivation kinetics of both bacteria were improved by the combination of the iron micronutrient with H_2O_2, showing the non-presence of a shoulder phase in the kinetic profile and reducing the treatment time required to achieve the DL in comparison with Fe^{3+}-EDDHA/solar, H_2O_2/solar and solar only disinfection processes. These results can be explained based on its higher oxidative capability and the more efficient and complex inactivation mechanism described elsewhere [16]. Briefly, this mechanism is based on accumulative damages on the bacteria cell membrane provoked

mainly by: i) the capability to generate •OH and other ROS by photo-Fenton reactions and ii) the permeability alteration of the cell membrane by the presence of the chelating agent [16].

Regarding reagent concentration, as for the Fe^{3+}-EDDHA/solar process, the disinfection efficiency was enhanced by increasing the iron concentration from 0.5 to 2.5 mg/L, while no improvement was observed for bacterial inactivation when increasing the concentration to 5 mg/L. A similar behavior was observed in a previous study of EDDS chelate, where the disinfection efficiency was improved by increasing the iron chelate concentration from 0.05 mM to 0.1 mM whereas a marked detrimental effect on the efficiency was observed when the concentration increase to 0.2 mM [27]. In other recent study that also use an aminopolycarboxylic acid (NTA, nitrilotriacetic acid) as iron chelate for the removal of three pharmaceutical micropollutants in continuous-flow mode, a significant removal improvement was not observed by using twice the concentration of reagents [33]. This behavior can be explained based on the combination of different reasons: i) the low iron quantity available, ii) the screen effect as consequence of iron precipitation [27] and iii) the increase of organic matter concentration coming from the organic chelate, a well-known competitor for the generated •OH radicals, decreasing the kinetic of radical-target reactions [34]. Therefore, to achieve the most effective treatment conditions, a compromise between these different factors is needed to obtain the optimal reagents concentration in CPC reactors.

In the present study, the SFCWW disinfection capability was markedly reduced at the iron chelate concentration of 0.5, attributed mainly to a reagent limitation at this concentration, where turbidity and organic matter detrimental effects of the sample prevailing over the solar treatment efficiency obtaining a similarly behavior when the iron chelate concentration rise up to 5 mg/L. Therefore, the faster kinetic inactivation was obtained with 2.5/20 mg/L of Fe^{3+}-EDDHA/H_2O_2 for both bacteria, where the DL was achieved after 60 min and 8.41 kJ/L of Q_{UV}. This treatment condition improved the efficiency of the other solar processes studied employing the same reagents concentrations separately (H_2O_2/solar: 20 mg/L; Fe^{3+}-EDDHA/solar: 2.5 mg/L). The Q_{UV} required for SFCWW disinfection by 2.5/20 mg/L of Fe^{3+}-EDDHA/H_2O_2 was 30 % and 78 % lower than the required for H_2O_2/solar (11.91 kJ/L) and Fe^{3+}-EDDHA/solar process (37.81 kJ/L), respectively.

In our previous work at laboratory scale (200 mL of volume), the Q_{UV} required to reach DL for both bacteria and similar reagent´s concentration was ca. 5 kJ/L [16], this value is very close to the 8.41 kJ/L obtained at pilot plant scale, but treating 60 L of contaminated SFCWW. This corroborates the possibility of up-scaling the Fe^{3+}-EDDHA/H_2O_2 for SFCWW disinfection using solar CPC reactor.

The residual iron in solution (Table 2, Figure SI2b) was significantly reduced compared with Fe^{3+}-EDDHA/solar, as only 12 to 25 % of the chelated iron initially added remain in solution at the end of the solar treatment for 0.5 to 5 mg/L of Fe^{3+}-EDDHA, respectively. In the case of the oxidant concentration (H_2O_2), it was almost completely removed during the treatment (90 %) in all the tested conditions (Figure SI2a), which was expected due to the oxidant is consumed during the photo-Fenton reactions to generate $^{\bullet}OH$. The low iron concentrations tested and the very low H_2O_2 residual concentration in the solar treated SFCWW makes possible a subsequent viable agricultural reuse without pose a risk to vegetables or irrigations systems [17, 35]. Moreover, the partial decomposition of the chelate during the solar treatment detected by the iron measurements (Fig. SI2b) and the UV-vis absorption profile of the treated water (Figure SI3) indicates that the chelate still remains active and available as source of iron for the further irrigation of vegetables, nevertheless it would require a new research to confirm it.

3.1.4 Post-treatment bacterial analysis

The efficiency of the solar processes was assessed in terms of bacterial regrowth after 24 h of dark storage. The concentration of *E. coli* O157:H7 and *S. enteritidis* detected after that period is shown in Figure 4. According to Spanish RD 1620/2007, the limit of *E. coli* concentration for wastewater irrigation is established at 100 CFU/100mL (RD 1620/2007) [21]. These results showed that after all the solar processes and reagent´s concentration tested, the bacterial concentration was less than this limit for both pathogens in the treated SFCWW, except for 2.5 mg/L of reagent in H_2O_2/solar process (for both pathogens) and 0.5/2.5 mg/L of Fe^{3+}-EDDHA/H_2O_2 (only for *E. coli* O157:H7). Therefore, the use of very low reagents concentrations (< 5 mg/L of H_2O_2 and 2.5 mg/L of Fe^{3+}-EDDHA) may not be appropriate to ensure efficient water disinfection and higher oxidative conditions should be required when application to real scenarios. On the other hand, considering the new European proposal for the regulation of wastewater reuse for irrigation, the presence of *E. coli* is more restrictive, as its concentration is limited to 10 CFU/100mL (Procedure 2018/0169/COD) [22]. For this regulation, the suitable solar processes for SFCWW disinfection and reuse in irrigation are H_2O_2/solar at concentrations higher than 10 mg/L of H_2O_2 and Fe^{3+}-EDDHA/H_2O_2 at 2.5/20 and 5/40 mg/L of reagents; while lower concentration as well as the Fe^{3+}-EDDHA/solar process can be discarded as appropriated treatments for SFCWW disinfection due to detection of *E. coli* at concentrations higher than this very restrictive value.

3.2 OMCs removal by solar processes

Simultaneously to the analysis of bacterial inactivation, the degradation of each OMC was also investigated in SFCWW. Figure 5 shows a comparison of the ΣOMCs removal profile obtained along the solar treatments, and the degradation of each individual OMC is shown in Figure SI4. In general, non-significant removal percentages were obtained by solar only treatment, due to the initial OMCs concentration remained almost constant during the treatment time, discarding therefore photolysis effects on the results obtained in this study. The percentage of ΣOMCs removal by H_2O_2/solar process at any reagent concentration tested varied from 10 to 20 %, which is a very modest efficiency. These low efficiencies were expected due to the high energy required for the cleavage of the O-O bond into ·OH and therefore, their dissociation will be generated only under shortwave wavelengths (< 290 nm) which are in a very small extent in the solar spectrum. The removal percentage obtained is in line with other study in literature, where degradation of ca. 20 % was reported for the antibiotic chloramphenicol by H_2O_2/solar process when it was exposed to similar accumulative energy (60 kJ/L) [36]. Although the Fe^{3+}-EDDHA/solar process has shown a disinfection capability, this process did not degraded OMCs at any of the Fe^{3+}-EDDHA concentrations tested (data not shown). The non-degradation observed are in agreement with the non-generation of oxidative species by this solar process.

The highest ΣOMCs degradation (42 %) of this study was attained with 2.5/20 mg/L of Fe^{3+}-EDDHA/H_2O_2, attributed mainly to the capability of ·OH generation of this process by photo-Fenton reactions [16]. Significantly, the OMCs degradation profiles (Figure 5) observed with this solar process in the two most oxidant conditions were non-linear and characterized by a fast degradation in the first stage of the process followed by a smooth decay until the end of the treatment time. The low chelate concentration employed and its self-degradation by the generated ·OH might explain the low efficiencies and the double kinetic degradation observed, behavior previously reported for other chelate agents like EDDS in ultrapure water [37] and DPTA (Diethylene triamine pentaacetic acid) or EDTA in wastewater effluents after the conventional activated sludge treatment [38]. In addition, DOC content was measured throughout all solar processes. The results indicated a very slight DOC degradation (lower than 10%) for both, H_2O_2/solar and Fe^{3+}-EDDHA/H_2O_2/solar process (data not shown).

In summary, as for disinfection, the highest ΣOMCs removal efficiency was also attained by the Fe^{3+}-EDDHA/H_2O_2/solar process using 2.5 and 20 mg/L of iron micronutrient and oxidant as Fenton reagents, respectively.

3.3 Toxicity evaluation

Despite the fact that phytotoxicity of the Fe^{3+}-EDDHA complex as fertilizer has been previously studied [39], as far as the author´s knowledge there are no studies dealing with the ecotoxicity of this chelating agent in wastewater samples.

Figure 6 shows the *A. fischeri* bioluminescence inhibition of SFCWW samples treated by the best operational conditions of the three solar processes investigated, i.e., H_2O_2/solar: 20 mg/L, Fe^{3+}-EDDHA/solar: 2.5 mg/L and Fe^{3+}-EDDHA/H_2O_2/solar: 2.5/20 mg/L. Additionally, two control tests (effect of the water matrix and Fe^{3+}-EDDHA at 2.5 mg/L) were also carried out.

A slight increase in the bioluminescence inhibition percentage (BI %) by the mere presence of Fe^{3+}-EDDHA was observed (from 12.4±5.7 to 21±6.1 %). This indicates that the presence of the commercial iron-chelate or any sub-product generated during its synthesis, somehow affected *A. fischeri* metabolism, not being possible to discard the effect of each one. Nevertheless, no significant toxicity by any treatments towards *A. fischeri* was obtained as lower BI than 50 % was detected in all cases, including control and solar treated samples.

For the H_2O_2/solar process, the luminescence emitted by the marine bacteria was not significantly affected (16.5 ± 3.5 BI %), whereas for the other two solar processes (Fe^{3+}-EDDHA/solar and EDDHA/H_2O_2/solar process) a BI % increase was obtained: 39.3 ± 3.1 and 24.5 ± 3 BI %, respectively. The higher BI % observed for the Fe^{3+}-EDDHA/solar process could be related with the presence of photodegradation products such as salicylaldehide, salicylic acid, salicylaldehyde ethylenediamine diimine or similar subproducts generated during the solar process [32]. Some of them have been reported to be harmful to aquatic microorganisms [40]. However, the lower BI % observed for Fe^{3+}-EDDHA/H_2O_2/solar process could be explained by the higher oxidative capability of this process (generation of $^{\bullet}OH$) which accelerates the decomposition of both the iron chelate and probably also their photodegradation products, therefore reducing the number of agents that can affect bacteria´s metabolism and reinforced the benefits of the application of this solar process for SFCWW treatment.

3.4 Techno-economic assessment

An economic analysis of the disinfection alone and the simultaneous disinfection and decontamination of SFCWW was carried out for the solar processes studied at the best operational conditions obtained for each one. For the costs estimation of simultaneous disinfection and decontamination, the accumulative UVA energy (Q_{UV}) obtained after 2 h of treatment was considered, taking into account that kinetics of OMCs degradation did not lead

to a significant improvement from 2 to 5 h of treatment time (Figure 5). The total treatment cost was calculated as a summation of the investment (IC) and operational cost (OP) based on the calculation of their annual cost.

The annual IC was estimated based on the cost of the CPC field required for each solar process, which represents the main investment cost for solar-driven systems, and the application of an 8 % of capital recovery factor (5 % of interest rate and 20 years of equipment life-time). The CPC field (A_{CPC}, m^2), was calculated according to the Eq. 6 [11]:

$$A_{CPC} = \frac{Q_{UV} \cdot V_{tot}}{T_s \cdot UV_G} \qquad \text{Eq. 6}$$

Taking into account the following assumptions:

- The Q_{UV} values obtained for water disinfection (in J/L): 11.91 x 10^3, 37.81 x 10^3 and 8.41 x 10^3 J/L for H_2O_2/solar (20 mg/L), Fe^{3+}-EDDHA/solar (2.5 mg/L) and Fe^{3+}-EDDHA/H_2O_2/solar (2.5/20 mg/L), respectively, considering the inactivation kinetics of *S. enteritidis* (the more resistant pathogen).
- The Q_{UV} values considered for the simultaneous disinfection and OMCs removal (in J/L): 25.24 x 10^3 and 17.03 x 10^3 J/L for H_2O_2/solar and Fe^{3+}-EDDHA/H_2O_2/solar, respectively.
- The annual volume of treated water (V_{tot}, L): 18250 x 10^3 L/year. Considering 50 m^3 per day and 365 operation days per year.
- Yearly time of operation (T_s, s): 157.68 x 10^5 s, 12 h of operation per day.
- The average of local solar UVA radiation (UV_G, W/m): 36.8 W/m^2.

As the rest of the equation parameters are constants, the Q_{UV} value for each process will be the parameter that determines the CPC field-area in each case being its value directly proportional to the necessary CPC area. The CPC area needed for SFCWW disinfection are: Fe^{3+}-EDDHA/solar (1189.2 m^2) > H_2O_2/solar (374.6 m^2) > Fe^{3+}-EDDHA/ H_2O_2/solar (264.5 m^2). For the simultaneous OMCs removal: H_2O_2/solar (793.8 m^2) > Fe^{3+}-EDDHA/ H_2O_2/solar (535.6 m^2). The final CPC cost per m^2 was calculated based on the estimation of different scaling-up prices: 816 €/m^2 for A_{CPC} < 1000 m^2 and 418 €/m^2 for A_{CPC} > 1000 m^2.

The operational costs were calculated based on the reagent, electricity and maintenance costs. The maintenance cost was considered to be 2.5 % of the annual IC. For the reagents cost estimation, only the H_2O_2 (industrial grade price of 0.43 €/L for a 35 % (w/v) solution) was

taken into account because the iron micronutrient was considered an agriculture cost (farmer) due to this iron chelate is usually added to the irrigation water by the farmers to avoid the iron chlorosis disease in arid or semiarid regions and therefore its addition would not entail any extra cost to carry out the solar treatment if its objective is the subsequent water reuse. Electricity costs were estimated considering a price of 0.155 € kW/h in Spain and the power required for two water centrifugal pumps: one for filling the reactor (0.22 kWh/m^3) and another one for water recirculation during the treatment time (0.44 kWh/m^3).

The estimated total costs to treat a m^3 of SFCWW by the three solar processes under study and a breakdown of the relative contribution of each cost are presented in Table 3. Non-significant differences were observed for the OC of each solar process (0.15-0.20 €/m^3) and therefore the IC was the responsible of the different costs estimated for each process representing < 85 and < 90 % of the total cost for disinfection alone and simultaneous disinfection and decontamination, respectively. The Fe^{3+}-EDDHA/H_2O_2/solar process showed, as we expected, the lower estimated IC (0.95 and 1.92 €/m^3) due to the higher efficiency of this solar process for the removal of the two types of targets (lower Q_{UV} value and therefore lower CPC field-area). The estimated price to treat one m^3 of SFCWW by the Fe^{3+}-EDDHA/H_2O_2/solar process were 1.10 € for disinfection alone and 2.10 € for the simultaneous disinfection and decontamination. This cost is 36 and 45 % lower than the estimated for SFCWW disinfection and OMCs removal by the H_2O_2/solar process, respectively.

The cost obtained in this study can be explained due to the water matrix complexity (100 NTU of turbidity) and the low water volume intended to be treated in this particular agro-food industry, which significantly may increase the investment cost based on scaling-up rules.

Regarding other technical aspects, there are several advantages of the use of this commercial iron-chelate respect to other commercial agents, such as EDDS, including: i) the photo-Fenton process using Fe^{3+}-EDDHA has showed to be able to improve the disinfection efficiency respect to its not addition (H_2O_2/solar process), ii) the high stability of this iron chelate gives to a higher 'lifetime' in solution than other chelating agents and lead to the presence of a residual iron chelate concentration in the solar treated wastewater, which allows a subsequent agricultural reuse in arid or semi-arid regions, contributing at the same time to solve two important problems in this regions: iron chlorosis disease of crops and water scarcity and, iii) among the different iron chelating agents authorized for agricultural use, EDDHA is the most used by the farmers (56-79% are EDDHA chelates) due to it is the most efficient (high stability in a wide range of pH) and therefore, the fact that its already use by the farmers (normally

added to the irrigation water) could contribute to the feasible real implementation of the proposed processes.

Finally, it can be also highlighted that the integration of these solar processes in the fresh cut-industry can be considered as a feasible and promising strategy to decrease its water footprint due to several reasons: i) the successful disinfection performance of the solar processes evaluated, ii) the low volume of wastewater generated by this particular agro-food industry (up to 50 m^3 per day) iii) the modular design of CPC reactors and the small area (ca. 250 m^2) required to treat such affordable amount of wastewater, iv) the clear benefits of irrigation of vegetables with a regenerated water containing potential substances (such as the iron-chelate and other organic substances and nutrients) that promote enhanced yields of the vegetables production, reducing at the same time some cultivation costs for farmers and, v) the easily adaptation of the proposed solar processes with the modular CPC reactor design for irrigation due to this industries are commonly located closely to the cultivation fields, reducing therefore the requirements related with water storages and distribution systems

4. CONCLUSIONS

The three solar processes studied (H_2O_2/solar, Fe^{3+}-EDDHA/solar and Fe^{3+}-EDDHA/H_2O_2/solar) has shown to disinfect successfully a water matrix with a high turbidity content (100 NTU) at pilot plant scale for the first time.

The Fe^{3+}-EDDHA/H_2O_2/solar process using low reagents concentrations (2.5/20 mg/L of Fe^{3+}-EDDHA/H_2O_2) has shown the highest treatment capability reducing ca. 40 % the OMCs load and attaining > 5-log reduction value for both pathogens with an accumulated solar energy of 8.41 kJ/L or 60 min of solar treatment time.

The high disinfection efficiency obtained by the three solar processes at the best operational conditions (H_2O_2/solar: 20 mg/L; Fe^{3+}-EDDHA/solar: 2.5 mg/L and Fe^{3+}-EDDHA/ H_2O_2/solar: 2.5/20 mg/L) accomplished the microbiological quality established in urban wastewater reuse law, including the Spanish Royal decree 1620/2007 (<100 CFU/100mL). Moreover, H_2O_2/solar and Fe^{3+}-EDDHA/H_2O_2/solar processes also fitted values of the new European proposal (Procedure 2018/0169/COD) (<10 CFU/100mL). These results have significant implications due to their capability of enabling the intended treated wastewater reuse for irrigation in agriculture with the incorporation of the iron micronutrient as an advantage.

608 The ecotoxicity findings suggest that the SFCWW treated by solar processes might not affect
609 the ecosystem's health. Nevertheless, this study was based on the toxic effect of only one
610 organism and more research using different tests should be performed to know the potential
611 toxic environmental effect of the solar treated water.
612
613 The estimated treatment cost obtained are high to be applied at industrial scale, but the
614 possibility of reusing the treated SFCWW directly for irrigation with an iron micronutrient that
615 farmers already used could contribute to the reduction of the water footprint in the fresh-cut
616 industry and the reduction of water scarcity and iron chlorosis in arid or semi-arid regions,
617 making this solar treatment a promising option.
618

619 ## 5. ACKNOWLEDGMENTS

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624

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774

Captions

Table 1. Bacterial inactivation kinetic constants.

Table 2. Initial and final reagent´s concentrations measured in SFCWW treated by all solar processes.

Table 3. Techno-economic data of solar treatments.

Figure 1. *E. coli* O157:H7 (a) and *S. enteritidis* (b) inactivation by H_2O_2/solar**.**

Figure 2. *E. coli* O157:H7 (a) and *S. enteritidis* (b) inactivation by Fe^{3+}-EDDHA/solar.

Figure 3. *E. coli* O157:H7 (a) and *S. enteritidis* (b) inactivation by Fe^{3+}-EDDHA/ H_2O_2/solar.

Figure 4. Analysis of bacteria concentration in treated SFCWW after 24 h of storage.

Figure 5. Degradation profiles of ΣOMCs by H_2O_2/solar and Fe^{3+}-EDDHA/H_2O_2/solar processes.

Figure 6. Ecotoxicity detected by *A. fischeri* test in untreated and solar treated SFCWW samples.

Table 1.

		E. coli O157:H7				*S. enteritidis*			
Treatment	**[Fe^{3+}:H_2O_2] (mg/L)**	**k (L/kJ)**	**R^2**	**SL (kJ/L)**	**Q_{UV} (kJ/L)**	**k (L/kJ)**	**R^2**	**SL (kJ/L)**	**Q_{UV} (kJ/L)**
Fig. 1a/1b									
Solar photo-inactivation*	-	0.529±0.104	0.832	-	56.56	0.243±0.011	0.983	1.57	56.56
H_2O_2/solar	2.5	0.400±0.038	0.947	-	13.60	k_1:0.413±0.043 k_2:0.065±0.005	0.948 0.979	-	25.20
	5	0.495±0.028	0.984	-	11.52	k_1:0.623±0.063 k_2:0.068±0.015	0.970 0.768	-	26.46
	10	0.510±0.103	0.796	-	11.60	k_1:0.543±0.061 k_2:0.062±0.020	0.940 0.754	-	17.05
	20	**0.626±0.105**	**0.920**	**-**	**8.75**	**0.429±0.040**	**0.965**	**-**	**11.91**
	40	1.099±0.203	0.825	-	4.89	0.554±0.040	0.954	-	10.34
Fig. 2a/2b									
Fe^{3+}-EDDHA/solar	0.5	k_1:0.331±0.078 k_2:0.031±0.002	0.850 0.960	8.23	56.82	0.090±0.006	0.949	3.78	56.82
	2.5	**0.417±0.026**	**0.992**	**19.12**	**31.41**	**0.229±0.029**	**0.939**	**13.51**	**37.81**
	5	0.244±0.026	0.954	17.05	40.78	0.191±0.022	0.938	17.05	47.11
Fig. 3a/3b									
Fe^{3+}-EDDHA/H_2O_2/solar	0.5:2.5	0.195±0.021	0.881	-	30.82	0.142±0.015	0.856	-	43.67
	2.5:20	**0.706±0.093**	**0.876**	**-**	**8.41**	**0.673±0.082**	**0.893**	**-**	**8.41**
	5:40	0.805±0.131	0.840	-	6.33	0.527±0.022	0.984	-	11.35

*Inactivation kinetic according to model 4, where residual bacteria population (N_{res}) for *E. coli* and *S. enteritidis* was 4 and 50 CFU/mL respectively.

Table 2.

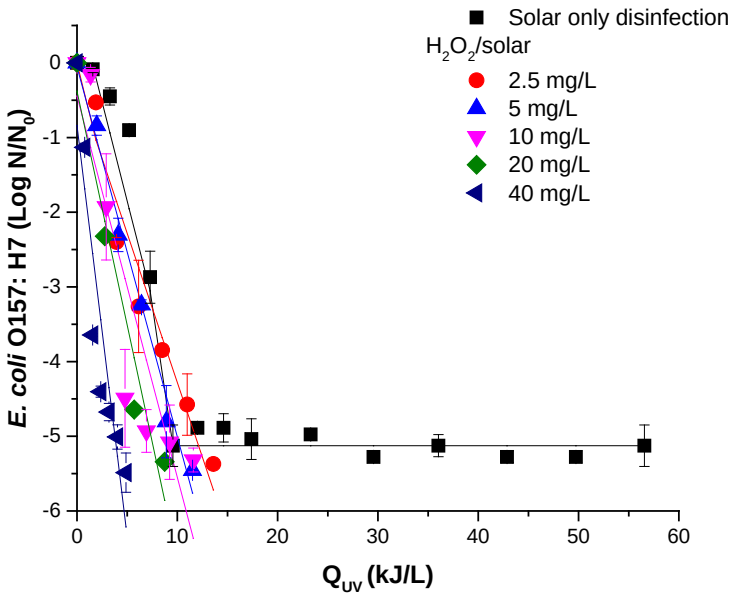
Treatment	[Fe:H_2O_2] (mg/L)	$[Fe]_{t300}$ (mg/L)	% residual	$[H_2O_2]_{t300}$ (mg/L)	% residual
Solar photo-inactivation	-	-	-	-	-
H_2O_2/solar	2.5	-	-	1.66	66
	5	-	-	3.13	63
	10	-	-	7.05	71
	20	-	-	14.30	72
	40	-	-	30.73	77
Fe^{3+}-EDDHA/solar	0.5	0.15	30	-	-
	2.5	0.98	39	-	-
	5	2.64	53	-	-
Fe^{3+}-EDDHA/H_2O_2/solar	0.5:2.5	0.06	12	0.46	9*
	2.5:20	0.42	17	1.66	8
	5:40	1.27	25	2.59	7

*Addition of 2.5 mg/L of H_2O_2 at 120 min

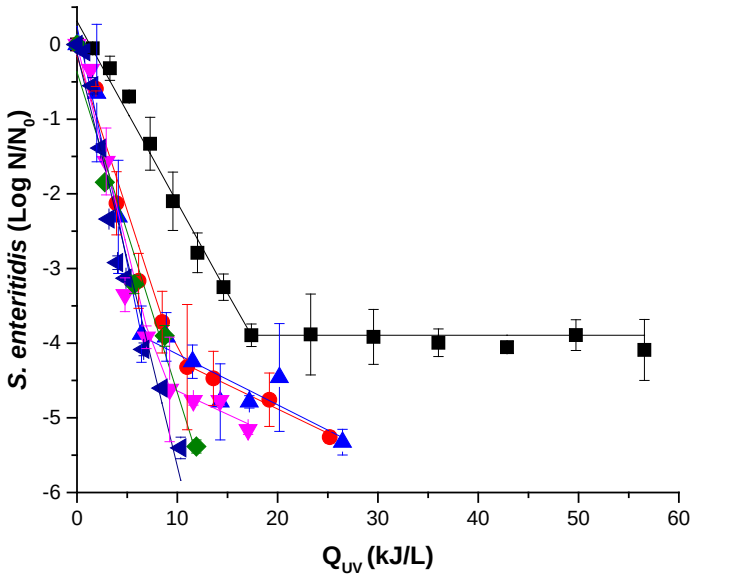
Table 3.

	Fe^{3+}-EDDHA/solar		**H_2O_2/solar**		**Fe^{3+}-EDDHA/H_2O_2/solar**	
	€/m^3	%	€/m^3	%	€/m^3	%
Disinfection						
IC	2.18	94	1.34	89	0.95	86
OC						
Electricity	0.10	4	0.10	7	0.10	9
Reagent	-	-	0.03	2	0.03	3
Maintenance	0.05	2	0.03	2	0.02	2
Total (€/m^3)	**2.33**		**1.50**		**1.10**	
Disinfection + OMCs						
IC	-	-	2.84	94	1.92	92
OC						
Electricity	-	-	0.10	3	0.10	5
Reagent	-	-	0.03	1	0.03	1
Maintenance	-	-	0.07	2	0.05	2
Total (€/m^3)	-	-	**3.04**		**2.10**	

Figure 1.

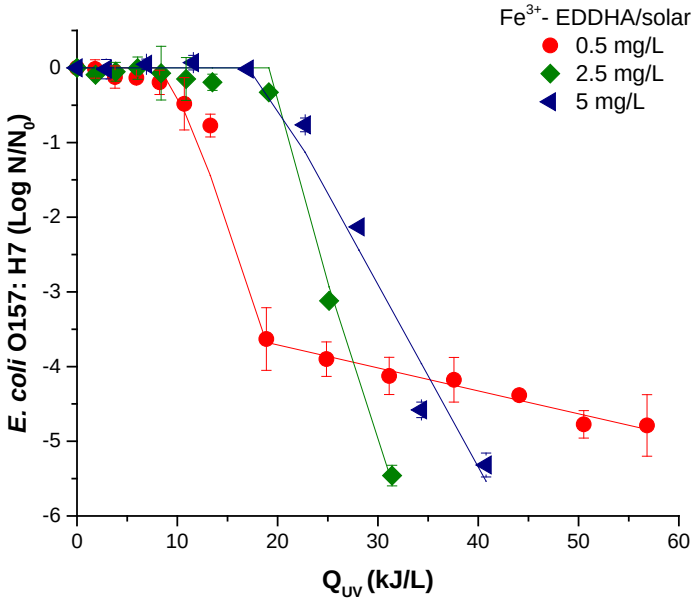

Solar only disinfection
H2O2/solar
2.5 mg/L
5 mg/L
10 mg/L
20 mg/L
40 mg/L
E. coli O157: H7 (Log N/N0)
QUV (kJ/L)

a)

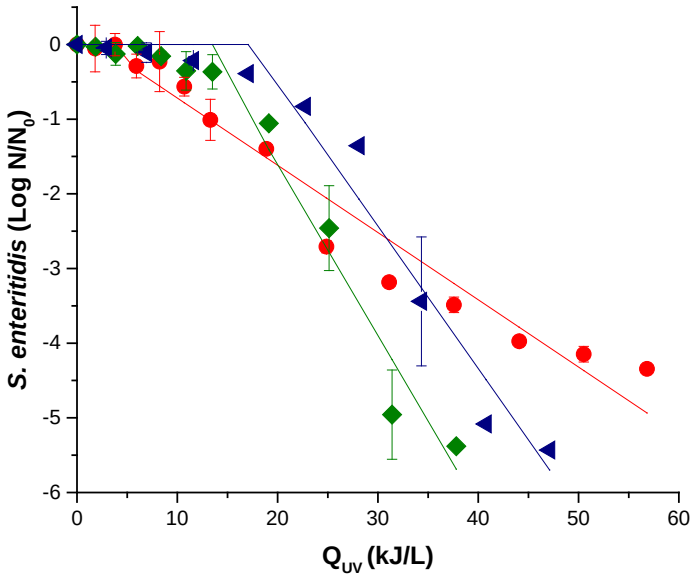

S. enteritidis (Log N/N0)
QUV (kJ/L)

b)

Figure 2.

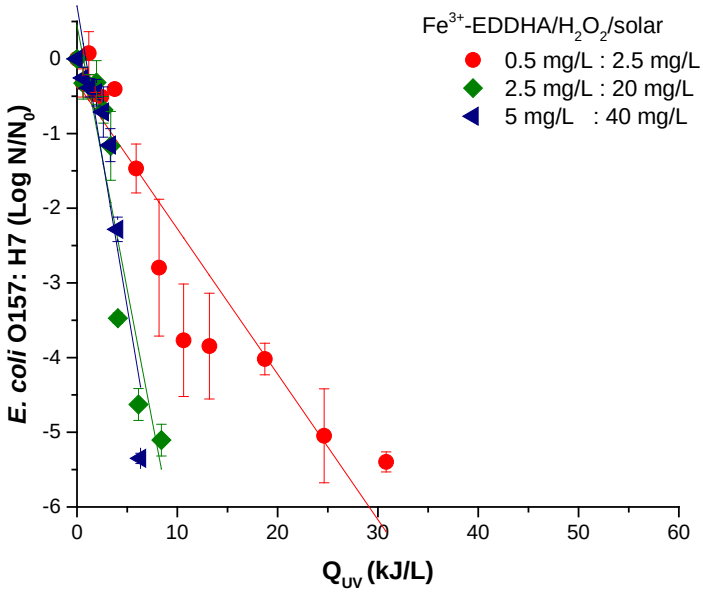

Fe3+- EDDHA/solar
0.5 mg/L
2.5 mg/L
5 mg/L
E. coli O157: H7 (Log N/N0)
QUV (kJ/L)
0
-1
-2
-3
-4
-5
-6
0
10
20
30
40
50
60

a)

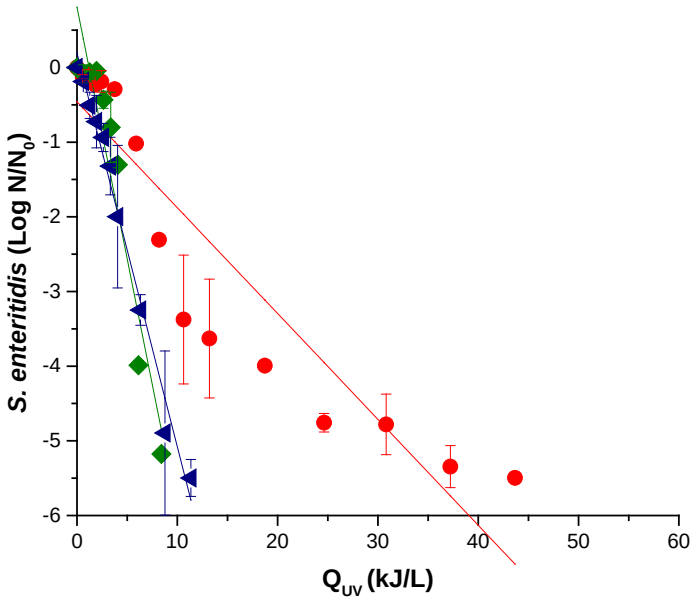

S. enteritidis (Log N/N0)
QUV (kJ/L)
0
-1
-2
-3
-4
-5
-6
0
10
20
30
40
50
60

b)

Figure 3.


Fe3+-EDDHA/H2O2/solar
0.5 mg/L : 2.5 mg/L
2.5 mg/L : 20 mg/L
5 mg/L : 40 mg/L
E. coli O157: H7 (Log N/N0)
QUV (kJ/L)
0
-1
-2
-3
-4
-5
-6
0
10
20
30
40
50
60

a)


S. enteritidis (Log N/N0)
QUV (kJ/L)
0
-1
-2
-3
-4
-5
-6
0
10
20
30
40
50
60

b)

Figure 4.

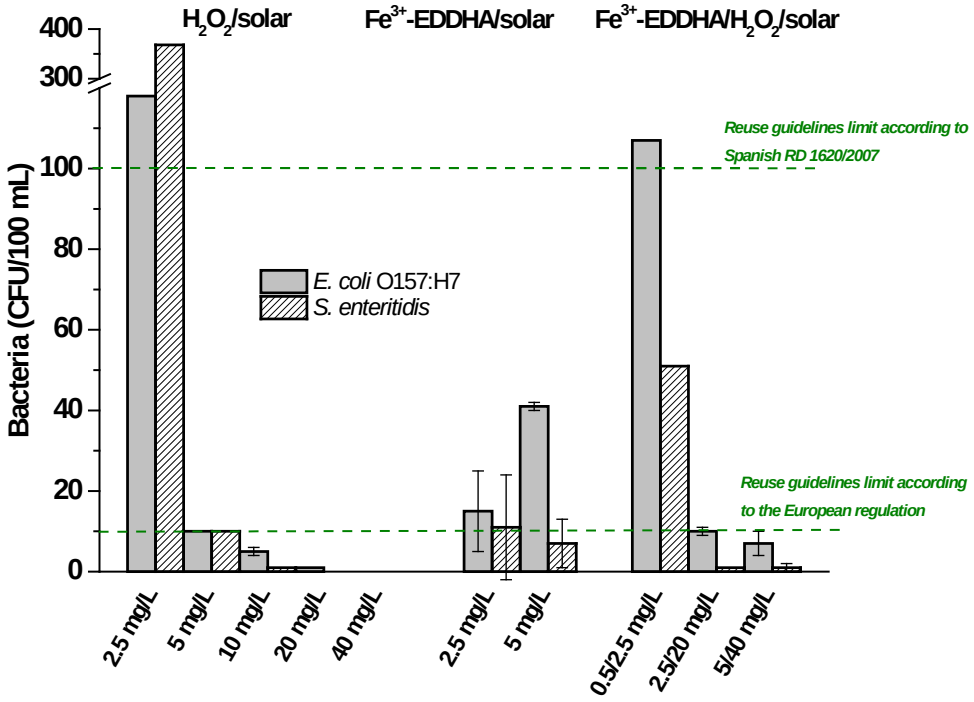

H2O2/solar
Fe3+-EDDHA/solar
Fe3+-EDDHA/H2O2/solar
Bacteria (CFU/100 mL)
400
300
100
80
60
40
20
0
Reuse guidelines limit according to Spanish RD 1620/2007
Reuse guidelines limit according to the European regulation
E. coli O157:H7
S. enteritidis
2.5 mg/L
5 mg/L
10 mg/L
20 mg/L
40 mg/L
2.5 mg/L
5 mg/L
0.5/2.5 mg/L
2.5/20 mg/L
5/40 mg/L

Figure 5.

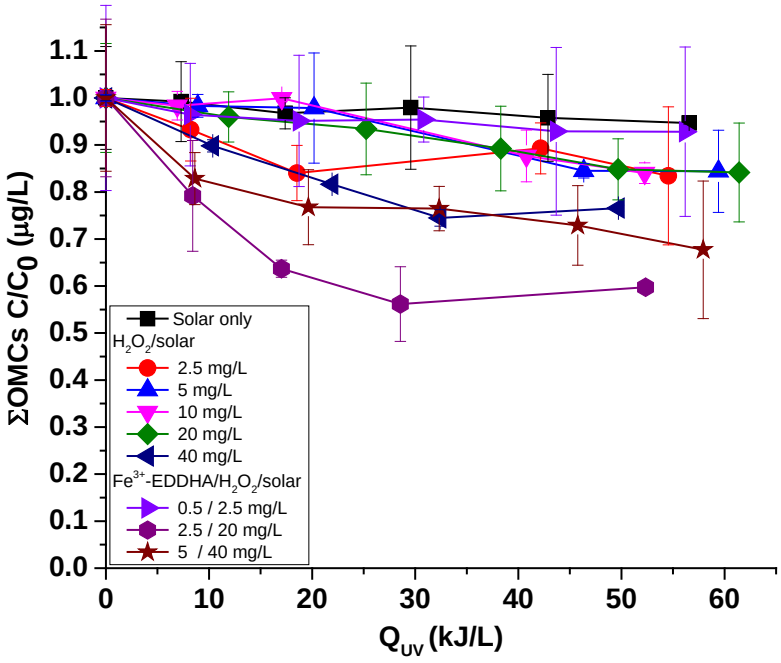
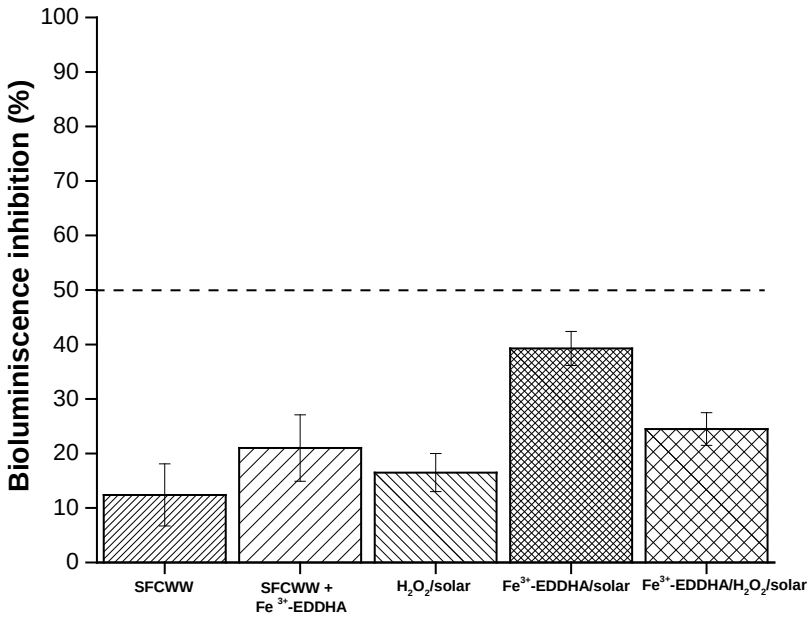

ΣOMCs C/C0 (μg/L)
QUV (kJ/L)
Solar only
H2O2/solar
2.5 mg/L
5 mg/L
10 mg/L
20 mg/L
40 mg/L
Fe3+-EDDHA/H2O2/solar
0.5 / 2.5 mg/L
2.5 / 20 mg/L
5 / 40 mg/L

Figure 6


Bioluminiscence inhibition (%)
100
90
80
70
60
50
40
30
20
10
0
SFCWW
SFCWW + Fe 3+-EDDHA
H2O2/solar
Fe3+-EDDHA/solar
Fe3+-EDDHA/H2O2/solar