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Disinfection of roof harvested rainwater inoculated with *E. coli* and Enterococcus and post-treatment bacterial regrowth: Conventional vs solar driven advanced oxidation processes

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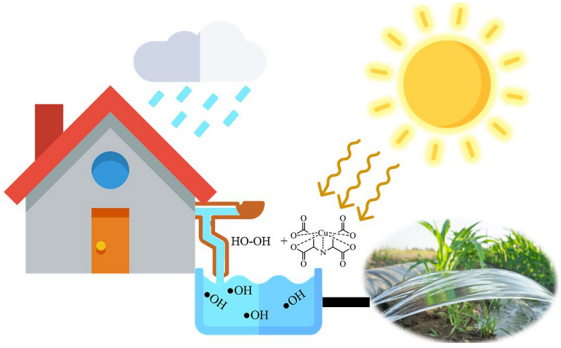
HIGHLIGHTS

- An alternative solar photo Fenton like process (SPF) was optimized for RHRW disinfection.
- All disinfection processes investigated inactivated *E. coli* faster than Entero.
- Sunlight/chlorine (SCL) was the most effective disinfection process (<15 min).
- UV-C radiation resulted in the higher bacterial regrowth (>50 CFU mL^{-1} after 48 h).
- SPF and SCL resulted in the lowest bacterial regrowth (<10 CFU mL^{-1} after 48 h).

GRAPHICAL ABSTRACT



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ABSTRACT

Solar driven advanced oxidation processes (AOPs) (an alternative solar photo Fenton like process (SPF), sunlight/H_2O_2 (SHP) and sunlight/chlorine (SCL)) and respective dark conditions, were compared for the first time to conventional (chlorination and UV-C radiation) disinfection processes, in the inactivation of *E. coli* and Entero strains inoculated in real roof-harvested rainwater (RHRW), to evaluate their possible safe use for crop irrigation. In this regard, bacterial regrowth was also evaluated 6, 12, 24 and 48 h after disinfection treatment. The SPF, using iminodisuccinic acid (IDS)-Cu complex as catalyst, was optimized (H_2O_2/IDS-Cu 55/1 best molar ratio) under mild conditions (spontaneous pH) and sunlight. The faster inactivation kinetics were observed for the SCL process ($k = 1.473\ min^{-1}$, $t_{1/2} = 0.47$ min for *E. coli* and $k = 1.193\ min^{-1}$, $t_{1/2} = 0.57$ min for Entero), while the most effective processes in controlling bacterial regrowth were SPF and SCL. Although UV-C radiation (0–$1.3 \times 10^4\ \mu W\ s\ cm^{-2}$ dose range) was the second faster disinfection process ($k = 1.242\ min^{-1}$, $t_{1/2} = 0.55$ min for *E. coli* and $k = 1.150\ min^{-1}$, $t_{1/2} = 0.60$ min for Entero), it was the less effective process in controlling bacterial regrowth (>10 CFU 100 mL^{-1} already after 6 h post-treatment incubation). According to the bacterial inactivation and regrowth tests carried out in this work, SPF and SCL are interesting options for RHRW disinfection, in case of effluent use for crop irrigation.

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1. Introduction

The latest World Water Development Report (2020) (Unesco et al., 2020) warns about the climate change effects on the water scarcity with consequences on water resources availability and an increase of the differences in the quality of life of millions of people all over the world. Water use has increased sixfold over the past century and it is rising by about 1% per year. Thus, the search for alternative water sources not supplied from fresh surface water or groundwater is strongly encouraged. Rainwater harvesting for domestic and agricultural uses is a very old practice dating back to 4500 BCE in the Middle East and India (Lofrano and Brown, 2010). In recent years, roof-harvested rainwater (RHRW) has drawing increasing attention as one of the most important alternatives to lessen the impacts associated with the uncontrolled use of potable water in many countries (Ali et al., 2020; Şahin and Manioğlu, 2019; Semaan et al., 2020). The replacement of drinking water with RHRW represents a great challenge mainly for a variety of non-potable uses. However, it is important to be aware of the risks associated with its use. Besides conventional contaminants, several pathogens such as *Legionella pneumophila*, *Nontuberculous mycobacteria*, *Pseudomonas aeruginosa*, and *Acanthamoeba* spp., (Ahmed et al., 2010; Albrechtsen, 2002; Hamilton et al., 2016) total and faecal indicator bacteria (FIB) such as *Escherichia coli* (*E. coli*) and *Enterococcus* sp. (Entero) have been detected in RHRW (Hamilton et al., 2016; Lee et al., 2012). *E. coli* is the FIB of choice for most countries that have irrigation water quality guidelines or regulations for food safety purposes (Uyttendaele et al., 2015). *E. coli* are commonly found in the feces of warm-blooded animals in high numbers (Ahmed et al., 2015). Bacterial concentrations can reach up to 10^3–10^4 CFU 100 mL^{-1} in RHRW (Ahmed et al., 2014; Hamilton et al., 2016; Islam et al., 2011; Leong et al., 2017). Primary treatment methods employed for the disinfection of RHRW include solar pasteurization (SOPAS) and solar disinfection (SODIS), amongst other techniques (McGuigan et al., 2012; Nalwanga et al., 2018). Nevertheless, it has been shown that the antioxidant systems activated by SODIS are UV sensitive and the inactivation strongly depends on the dose of UV-A radiation (Kapuscinski and Mitchell, 1981). In addition, an increased turbidity (Nephelometric Turbidity Units, NTU > 30) of the water may also influence the efficiency of this system affecting the direct penetration of UV radiation (Dawney and Pearce, 2012; Dunlop et al., 2011). Chlorination is the most common disinfection method because it is cheap and effective in the inactivation of microorganisms. However, the formation of hazardous chlorination by-products (DBPs), is pushing towards alternative treatments, including advanced oxidation processes (AOPs) which present excellent capability to inactivate waterborne pathogens through highly reactive oxidizing species (Fiorentino et al., 2017; O'Dowd and Pillai, 2020; Ying Liu et al., 2020; Rizzo et al., 2020). Moreover, AOPs seem to be more effective than chlorination to reduce bacterial regrowth after disinfection (Fiorentino et al., 2015), which is a primary issue in RHRW and wastewater reuse practices, taking into account that they may be stored for hours/days before use. Regrowth tests are not only useful to understand possible risk related to the reuse of treated water for crop irrigation, but also to better evaluate the effect of disinfection processes on bacteria inactivation (Di Cesare et al., 2020a). Hydrogen peroxide (H_2O_2) and the Fenton reagents have been used to enhance SODIS processes, reducing the exposure times and controlling bacterial regrowth (Feng et al., 2020). Traditionally Fenton and photo-Fenton processes are carried out at acidic pH. However, when applied to RHRW treatment this condition presents several drawbacks, including costs related to pH conditioning. Thus, to run photo-Fenton treatment at mild conditions, Fe^{2+} is replaced by other metals (namely Cu^{2+}, Mn^{2+}, Co^{2+}, and Ag^{+}), possibly combined to organic or inorganic ligands to form complexes and/or to stabilize the metals over a wide pH range (Fiorentino et al., 2018; Prete et al., 2021; Wang et al., 2016). The UV/chlorine process has received an increasing attention as alternative AOP for the degradation of a number of pollutants (Cerreta et al., 2020; Miklos et al., 2019; Sgroi et al., 2021), but only a few studies have been focused on sunlight/chlorine and its effect on microorganisms inactivation under realistic conditions (Zhou et al., 2014).

The application of AOPs for the disinfection of RHRW has been poorly investigated so far (Campisano et al., 2017) and it does not exist, to authors' knowledge, any comparative investigation between conventional (e.g., chlorination and UV-C radiation) and non-conventional (e.g., AOPs) disinfection processes.

In this study conventional disinfection processes, namely chlorine and UV-C radiation, and solar-driven AOPs were comparatively investigated for the first time in RHRW disinfection to evaluate their possible safe use for crop irrigation. Solar driven AOPs included an alternative solar photo Fenton like process (SPF) working under mild conditions, using iminodisuccinic acid (IDS)-Cu complex as catalyst, sunlight/H_2O_2 (SHP) and sunlight/chlorine (SCL). Their effects were evaluated on *E. coli* and Entero strains, purposely selected from real RHRW, in terms of bacterial inactivation and regrowth. SPF process with IDS-Cu has not yet been investigated in the inactivation of bacteria in RHRW, being previously applied to secondary treated urban wastewater and found to be more effective than conventional Fenton and photo Fenton processes (Di Cesare et al., 2020a, 2020b; Fiorentino et al., 2018). In addition, it is the first time that this process is tested coupled to solar radiation.

2. Materials and methods

2.1. Chemicals

H_2O_2 at 35% w/w (Merck, Germany) was used in Fenton like and solar driven AOPs. Sodium hypochlorite (NaClO) solution at 10% (Sigma–Aldrich, St. Louis, MO) was used in chlorination and chlorine/sunlight tests. Cu-IDs was prepared according to the procedure reported in our previous work (Fiorentino et al., 2018), and used as catalyst in Fenton like and SPF like treatment. The IDS concentration used in the complex preparation was optimized in order to make negligible the scavenger effects.

0.1 ml for each 100 ml of samples of sodium thiosulfate 0.2 N, (Sigma–Aldrich, St. Louis, MO) and 2 ml for each 100 ml of samples of 2300 U mg^{-1} of bovine liver catalase at 0.1 g l^{-1} (Sigma-Aldrich, USA), were used to quench residual chlorine and H_2O_2 in chlorination and AOPs tests, respectively.

2.2. RHRW and selection of E. coli and Entero strains

RHRW samples were collected from the roof of a building in the Fisciano campus of University of Salerno (Italy), in three different days to carry out three different replicas. The RHRW was collected by a downpipe connected to the roof, in sterile containers of 2 l. Chemical and microbiological analysis were performed for each sample according to standard methods (APAT and Istituto di ricerca sulle acque, 2003). The collected samples were carried immediately in laboratory and stored at 4 °C. *E. coli* and Entero strains were selected from RHRW samples through membrane filtration and subsequent cultivation (24 h incubation time at 37 °C) on EC with MUG Fluorogenic Agar (Condalab, Madrid, Spain) and Enterococcus Selective Agar (Fluka, St. Louis, MO), respectively. Briefly, 100 ml of RHRW samples or its serial dilutions were filtrated through membrane and incubated on the corresponding selective agar. Some colonies were randomly picked up and frozen in 15% glycerol Triptone Soy Broth (TSB) at −20 °C for the subsequent disinfection experiments.

2.3. Inoculum and sample preparation

In order to operate under more controlled conditions and better investigate the inactivation kinetics, the RHRW was inoculated with the selected indigenous *E. coli* and Entero strains to reach 10^5 CFU

100 ml^{-1}. The selected *E. coli* and Entero were unfrozen and reactivated by streaking on respective selective agar and incubated at 37 °C for 24 h. Different colonies from the plate were inoculated into 14 m l sterile Tryptic Soy Broth (TSB, Sigma-Aldrich, St. Luois, MO) and incubated at 37 °C for 18 h by constant agitation in a rotator shaker to obtain a stationary phase culture. Cells were harvested by centrifugation at 3000 rpm for 10 min and the pellet was re-suspended in 14 mL phosphate buffer saline (PBS, Oxoid), yielding a final concentration of 10-^{9} CFU ml^{-1} approximately. An aliquot of this solution was added to the RHRW to achieve the desired initial bacterial load. Physic-chemical and microbiological average values of RHRW samples are included in Table 1.

2.4. Bacterial count

Bacterial count was performed by the membrane filtration method (APHA, 1998). Briefly, 10 ml of RHRW or its serial dilutions samples were filtered through 0.45 μm, pore size cellulose nitrate membranes (Millipore, Billerica, MA, USA), in according to Italian APAT-IRSA standard methods (APAT and Istituto di ricerca sulle acque, 2003), placed onto selective agar and incubated at 37 °C for 24 h and 48 h for *E. coli* and Entero, respectively. Measurements were carried out in triplicate and average values and standard deviation were plotted as CFU 100 ml^{-1}.

2.5. Disinfection tests

2.5.1. Fenton like and photo-Fenton tests

Preliminary tests, using *E. coli* as target strain, were carried out to find the optimum ratio between H_2O_2 and IDS-Cu as well as the optimum disinfectant doses in SPF and chemical disinfection processes, respectively. To assess the efficiency of the IDS-Cu complex as effective catalyst for Fenton like and SPF processes in RHRW treatment, different H_2O_2/IDS-Cu ratios (20, 35, 55, 75, 110, 165 H_2O_2/IDS-Cu molar ratio equivalent to 10, 20, 30, 40, 60 and 90 mg l^{-1} of H_2O_2 and 1 mg l^{-1} of Cu) were tested at different treatment times. In both processes the catalyst was added to the sample and after 2 min of stirring in the dark H_2O_2 was added. The Cu concentration was kept constant at 1 mg l^{-1} (corresponding at 0.015 mM) to not exceed the Cu concentration limit established by the Italian regulation for the wastewater reuse (*Decreto Legislativo 152/06 "Norme in materia ambientale,"* 2006). H_2O_2/IDS-Cu tests were carried out in a 500 mL cylindrical glass reactor (10.0 cm in diameter), completely covered with aluminum sheet and filled with the 500 ml of RHRW. H_2O_2/IDS-Cu /sunlight tests were carried out under natural sunlight irradiation at the Fisciano Campus of University of Salerno, located at 40° 46′ N and 14° 77′ E, in clear sunny days. Borosilicate glass bottles (DURAN, Schott, Germany) with a volume of 500 ml, magnetically stirred during all the experiment were exposed to sunlight, as described in a previous work (Fiorentino et al., 2015). After carrying out the preliminary tests to identify the best H_2O_2/IDS-Cu ratio both in dark and in sunlight conditions, H_2O_2 and H_2O_2/ sunlight tests as a comparison/control were conducted under the same conditions and with the same H_2O_2 concentrations used in the best H_2O_2/IDS-Cu and H_2O_2/IDS-Cu/sunlight tests. H_2O_2 residual was measured with the HR Chlorine Pocket Colorimeter (PCII) by DPD Chlorine method adapted by Hach. After sampling, catalase was added to RHRW samples in order to eliminate the residual H_2O_2. H_2O_2 and catalase at these concentrations do not have detrimental effects on *E. coli* viability (García-Fernández et al., 2012).

Table 1
Chemical characterization of RHRW (average values and standard deviation of six analyzed samples are reported).

Parameter	Value	Unit
pH	6.7 ± 0.2	
Conductivity	750 ± 35	(μS cm^{-1})
Turbidity	3.1 ± 0.5	(NTU)
TSS	15.0 ± 1.0	(mg l^{-1})
COD	40 ± 7	(mg l^{-1})
BOD	7 ± 5	(mg l^{-1})
Phosphate	1.1 ± 0.1	(mg l^{-1})
NO_2^-	1.2 ± 0.5	(mg l^{-1})
NO_3^-	8.5 ± 1.9	(mg l^{-1})
E. coli	750 ± 50	CFU 100 ml^{-1}
Entero	165 ± 15	CFU 100 ml^{-1}

2.5.2. Chlorination tests

Chlorination tests were carried out in dark and in presence of sunlight in the same operative conditions used for the other AOPs test. The concentrations of chorine used in these tests were 1, 2, 3, 4, 5, 7.5 and 10 mg l^{-1}. After sampling, 0.1 ml of sodium thiosulfate solution (10%) was added to each 100 ml sample to remove residual chlorine before bacterial count. Free residual chlorine was measured by a portable spectrophotometer (Pocket colorimeter Chlorine; Hach, Loveland, CO, USA).

2.5.3. UV-C radiation tests

UV-C experiments were carried out with a UV-C 16 W lamp (Sankyo Denky G10T5L, Japan) located in vertical position at the center of a 1.0 l glass cylindrical reactor (5.0 cm in diameter), completely covered by an aluminum sheet and filled in with 500 ml of RHRW. The UV-C reactor was placed in a water bath to keep the temperature at 25 °C during the experiments. The samples, continuously stirred, were exposed to a range of UV-C doses (0–1.3 × 10^4 μW s cm^{-2}) by varying the exposure time from 0 to 120 min. The corresponding UV-C dose was calculated by multiplying the irradiation time for the intensity of UV-C lamp measured at the bactericidal wavelength (i.e., 254 nm). All samples were analyzed in triplicate.

2.6. Bacterial regrowth tests

To evaluate the regrowth of *E. coli* and Entero bacterial cells in untreated (as control) and treated RHRW, samples were incubated at 20 °C for 12, 24 and 48 h. Briefly, 100 m l of RHRW or its serial dilutions samples were filtered through 0.45 μm pore size cellulose nitrate membranes. The samples for regrowth experiments were collected at the end (total bacterial inactivation) of disinfection experiments. Bacterial count was performed in triplicate.

2.7. Analytical instruments

All RHRW samples were characterized by using portable multiparameter pH-meter HI-98115 Hanna Instrument. The COD was measured by Varian Cary® UV-VIS spectrophotometer according to Li et al. (2009) method. Bicarbonates, phosphate, NO_2^-, NO_3^- were analyzed by multiparameter bench photometer HI83-200-02 Hanna. A Black Comet C 200-850 nm UV-VIS spectrometer (Stellar Net, Florida, USA) was used to measure the irradiance spectra of the UV lamp.

2.8. Kinetics analysis

Bacteria inactivation was described by pseudo-first-order kinetics, according to the Eq. (1)

$$-\ln\frac{C_t}{C_0} = kt \quad (1)$$

where C_t and C_0 are the concentrations of viable bacteria (CFU 100 ml^1) at time t and t_0, respectively; k is the pseudo-first-order rate constant of bacteria removal (min^{-1}) and t is the treatment time (min). k was determined by Data Analysis Software OriginLab 2018 (OriginLab Corporation 2018, Massachusetts, USA) by non-linear estimation (Least

Squares Estimation). Half-life time of bacteria inactivation ($t_{1/2}$, min), and the time for a 90% bacterial inactivation ($t_{90\%}$) were determined according to the Eqs. (2) and (3), respectively

$$t_{1_2} = \frac{ln\,2}{k} \tag{2}$$

$$t_{90\%} = \frac{2.3}{k} \tag{3}$$

The inactivation rate was calculated also as a function of both experimental time (t) and cumulative energy per unit of volume (Q_{UV}) as described in a previous work of Fiorentino et al. (2015). Solar radiation was measured in terms of incident UVA (in W m^{-2}), which is the radiant UVA energy rate incident on a surface per unit area. The average UVA irradiance for all tests was 23 ± 3 (standard deviation) W m^{-2} within the period 11:00–14:00 local time, with a minimum value of 18 W m^{-2} and a maximum value of 31 W m^{-2}. The inactivation kinetics were calculated as function of both experimental time (t) and cumulative energy per unit of volume (Q_{UV}) received in the photoreactor, and calculated by Eq. (4), while the graphs have been plotted as a function of time:

$$Q_{UVA,n} = Q_{UVA,n-1} + \Delta t_n UVA_{G,n} S_b / V_b \tag{4}$$

where $Q_{UVA,n}$, $Q_{UVA,n-1}$ are the UVA energy accumulated per liter (kJ l^{-1}) at times n and n−1, $UVA_{G,n}$ is the average incident radiation on the irradiated area, Δt_n is the experimental time of sampling, S_b is the illuminated area of the reactor (m^2), V_b is the total volume of treated RHRW (l). Q_{UVA} is generally used in order to compare results from tests operated under different weather conditions (Malato et al., 2009). The average Q_{UVA} measured during all experiments was 31 ± 6.7 W m^{-2}.

3. Results and discussion

3.1. Optimization of reagent doses for disinfection processes

The optimum molar ratio H_2O_2/IDS-Cu was found to be 55 in SPF test and 110 in Fenton test of H_2O_2 respectively and 1 mg l^{-1} of Cu (Fig. 1). Although the target contaminants (*E. coli* vs phenols) and aqueous matrices (RHRW vs deionized water) were different, the optimum H_2O_2/IDS-Cu ratio was found to be consistent with our previous work (Fiorentino et al., 2019).

Any *E. coli* removal could be observed in Fenton process before 40 min. After 90 min with 110/1 H_2O_2/IDS-Cu molar ratio, 4.7 log units removal was achieved and only a slight improvement could be observed after 180 min of treatment time (5 log units). In SPF a significant bacterial inactivation was observed after just 10 min treatment. With 55/1 H_2O_2/IDS-Cu molar ratio, 5 log units removal was obtained after 40 min. No significant variation could be observed by further increasing the treatment time up to 60 min. On the opposite with higher and lower molar ratio the bacterial inactivation increased up to 60 min of treatment (4.5 log units with 110/1 H_2O_2/IDS-Cu molar ratio and 3.5 log units with 75/1 H_2O_2/IDS-Cu molar ratio). In the Fenton tests, the concentration of residual H_2O_2 at the end of experiment was 55 ± 5, 25 ± 4, 24 ± 2, 12 ± 2, 2.5 ± 0.9, and 0.8 ± 0.1 mgl^{-1} for 165, 110, 75, 55, 35 and 20 H_2O_2/IDS-Cu molar ratios, respectively. In the SPF tests the concentration of residual H_2O_2 at the end of experiment was 31 ± 4, 3 ± 2, 0.17 ± 2.3, 0.05 ± 0.1, and 0 mg l^{-1} for 110, 75, 55, 35 and 20 H_2O_2/IDS-Cu molar ratios, respectively. Therefore, with the optimal molar ratio of 55 for the SPF process, the residual concentration of H_2O_2 was less than 0.20 mg l^{-1}, while with the optimal molar ratio of 110 for the Fenton process, the residual concentration of H_2O_2 was 25. ±4 mg l^{-1}. So far, no residual concentration of H_2O_2 has been established for wastewater reuse. However, it has been demonstrated that up to 50 mgl^{-1} of residual H_2O_2 concentration is not toxic for cultivated plants since the farmers use this reactant for maintaining

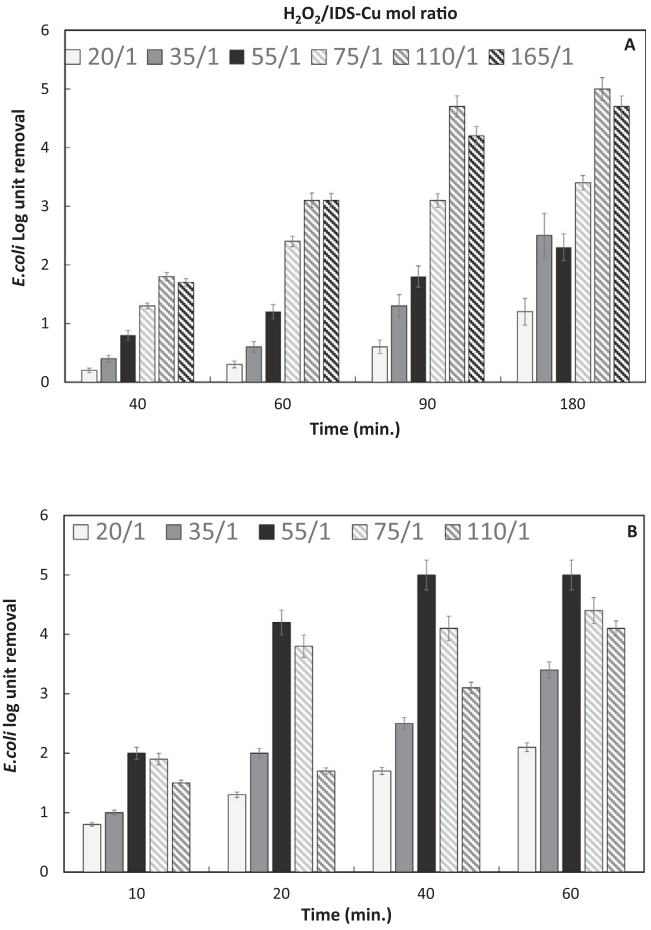

Fig. 1. *E. coli* log units inactivation by Fenton (H_2O_2/IDS-Cu) (A) and SPF (H_2O_2/IDS-Cu/sunlight) (B): optimization of H_2O_2/IDS-Cu mol ratio.

the crops disinfected (Chikthimmah et al., 2005). The very low residual oxidant concentration (less than of 0.2 mg l^{-1} of H_2O_2) in SPF tests allowed to exclude that even a minimal concentration of H_2O_2 could affect bacterial regrowth and to have a more reliable comparison with chlorination. Preliminary chlorination tests were carried out with different initial concentrations (from 1 to 10 mg l^{-1}) in order to obtain residual chlorine concentration at the end of experiment less than 0.2 mg l^{-1} to meet current Italian legislation for wastewater reuse (DM 185/2003). The initial concentration that allowed to get a residual active chlorine <0.2 mg l^{-1} (0.17 ± 0.06 and 0.07 ± 0.05 in chlorination and SCL processes, respectively) at the end of experiments was 3 mg L^{-1} of NaClO. Accordingly, all chlorination-based experiments were carried out with 3 mg l^{-1} of initial chlorine concentration. In all disinfection tests the pH of the solutions was around 6.7 ± 0.2 and dropped to a minimum of 6.3 ± 0.2. Under the investigated pH values, Cu is quantitatively solubilized as metal complex with IDS and no precipitation as hydroxide occurs. This result also highlights the high stability of the ID-Cu catalyst in an extended range of pH as reported in our previous work (Fiorentino et al., 2018). The optimized H_2O_2/IDS-Cu molar ratio reduces the hydroxyl radicals scavenging function of the IDS-Cu favoring the Fenton process with high productivity (Prete et al., 2021). Fiorentino et al. (2019) identified the best UV-C/H_2O_2/IDS-Cu molar ratio of 75 for phenol degradation; in this case the best condition 100 in the absence of radiation and 55 with solar radiation, are slightly different. This is possibly due to the different type of radiation used (with solar radiation the Cu-IDS seems to be more efficient and therefore the consumption of H2O2 increases) and different matrix, phenol being an aromatic substance more difficult to remove than bacteria. In SPF the best molar ratio of 55 is probably due to two factors. It is well known that high H_2O_2 residual concentration can

act as radicals scavenger (Huang et al., 2013). Accordingly, a further increase of H_2O_2 concentration (H_2O_2/IDS-Cu mol ratio 165/1) may explain the reduced process efficiency (Fig. 1A). Therefore, the determination of the optimum H_2O_2/IDS-,Cu mol ratio represents a crucial aspect for the optimization of the process. For a given complex concentration, a high H_2O_2 concentration can reduce the process efficiency due to radical scavenging effect. On the opposite, a lower H_2O_2 concentration can reduce the overall process efficiency. The text was revised accordingly.

3.2. Bacterial inactivation by different disinfection processes

The inactivation profiles of *E. coli* and Entero under optimal H_2O_2/IDS-Cu molar ratio, H_2O_2 disinfection and chlorination are plotted in Fig. 2. Spontaneous bacteria reduction without disinfection processes was also monitored as control. The inactivation of *E. coli* and Entero under optimal H_2O_2/IDS-Cu molar ratio for SPF, SHP, SCL and UV-C processes are plotted in Fig. 3. Based on the optimal molar ratio in Fenton and SPF, the H_2O_2 based processes (H_2O_2 and SHP) were carried out at 60 and 30 mg l^{-1} of H_2O_2. The kinetic bacteria inactivation values are shown in Table 2.

As expected, the initial bacterial load did not change during control test (dark condition without reagents). After 180 min, H_2O_2 resulted in a relatively low inactivation rate of 1.5 log units ($k = 0.076\ min^{-1}$, $t_{1/2} = 9.12$ min) and 1 log units ($k = 0.061\ min^{-1}$, $t_{1/2} = 11.34$ min) for *E. coli* and Entero, respectively and a residual H_2O_2 concentration of 47 ± 6 mg l^{-1} was detected. The limited consumption of H_2O_2 in the absence of a catalyst is confirmed in several works (Fiorentino et al., 2019; Mejri et al., 2020; Soriano-Molina et al., 2021). A complete inactivation *E. coli* ($k = 0.151\ min^{-1}$, $t_{1/2} = 4.58$ min) and Entero ($k = 0.093\ min^{-1}$, $t_{1/2} = 7.24$ min) by Fenton process, was observed after 120 and 180 min, respectively. The fastest inactivation kinetics were achieved by chlorine, where a completed inactivation for *E. coli* ($k = 1.134\ min^{-1}$, $t_{1/2} = 0.62$ min) and Entero ($k = 0.419\ min^{-1}$, $t_{1/2} = 1.65$ min) was observed after 20 and 30 min, respectively (Table 2). As described in the previous work of Di Cesare et al. (2020a), the bacterial inactivation generated by radical species using Cu^+ and Cu^{2+} cupper oxidation with H_2O_2 is similar to the Fe^{2+}/H_2O_2 and Fe^{3+}/H_2O_2 reaction systems (Eqs. (5) and (6)):

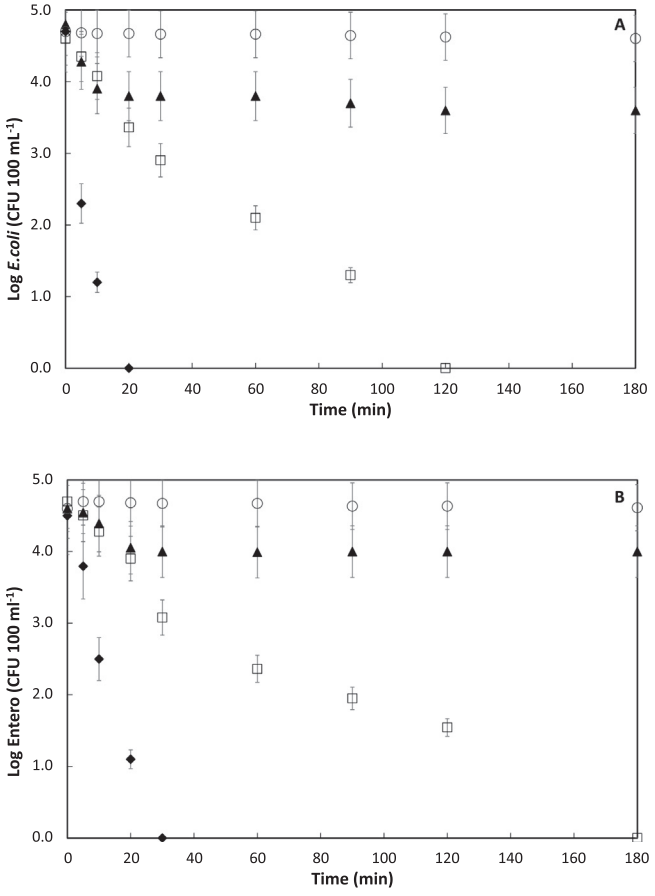

Fig. 2. Inactivation of *E. coli* (A) and Entero (B) with (▲) H_2O_2 60 mg l^{-1}, (♦) Chlorine 3 mg l^{-1}, (□) H_2O_2/IDS-Cu 55/1 mg l^{-1} and (○) control.

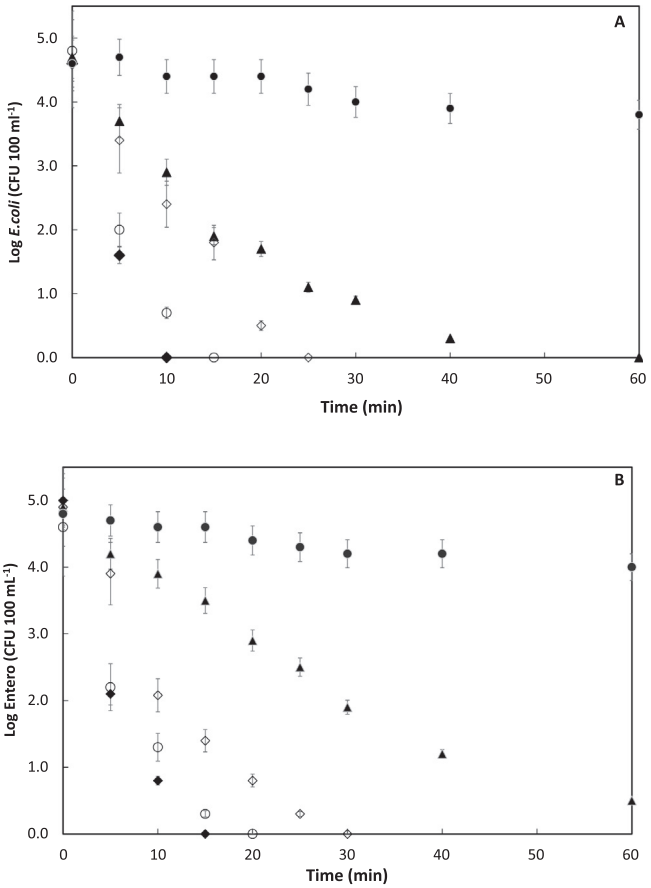

Fig. 3. Inactivation of *E. coli* (A) and Entero (b) with (○) UV-C, (▲) H_2O_2/sunlight 30 mg l^{-1}, (♦) chlorine/sunlight 3 mg l^{-1}, (◊) H_2O_2/IDS-Cu/sunlight 30/1 mg l^{-1} and (●) sunlight in the different disinfection test.

$$Cu^{2+} + H_2O_2 \rightarrow Cu^{+} + HO_2^{\bullet} + OH^{-} \quad \left(k = 4.6 \times 102\ M^{-1}\ s^{-1}\right) \tag{5}$$

$$Cu^{+} + H_2O_2 \rightarrow Cu^{2+} + HO^{\bullet} + HO^{-} \quad \left(k = 1.0 \times 104\ M^{-1}\ s^{-1}\right) \tag{6}$$

In all experiments. *E. coli* were inactivated faster than Entero. The higher resistance showed by these Gram-positive enteric bacteria against the disinfection treatments applied, compared to TC and *E. coli* (Gram-negative) could be attributed to the more resistant cell wall (García-Fernández et al., 2019; Giannakis et al., 2016). The cellular structure of these two bacterial species is different, in Entero (Gram-positive) the cell membrane (cytoplasmic lipid membrane), has a thicker peptidoglycan layer. In Gram-positive bacteria, up to 90% of the wall is peptidoglycan. Although some bacteria have only a single layer of peptidoglycan surrounding the cell, many Gram-positive bacteria have several sheets of peptidoglycan stacked on top of each other (Madigan et al., 2018). The peptidoglycan is thought to be deposited by the cell in 'cables' approximately 50 nm wide, with each cable consisting of several cross-linked glycan filaments. As the peptidoglycan 'matures', the cables themselves become cross-linked to form an even stronger cell wall structure. For this reason, usually Gram-positive

Table 2
Bacterial inactivation by disinfection test: kinetic parameters and Q_{UV} values corresponding to $t_{1/2}$ ($Q_{UV,t1/2}$) and to the time of 90% of total inactivation ($Q_{UV,\ t90\%}$).

	E. coli					Entero				
	R^2	k (min^{-1})	$t_{1/2;}$ $t_{90\%}$ (min)	$Q_{UV,t,1/2}$ (kJ l^{-1})	$Q_{UV,t,90\%}$ (kJ l^{-1})	R^2	k (min^{-1})	$t_{1/2;}$ $t_{90\%}$ (min)	$Q_{UV,t,1/2}$ (kJ l^{-1})	$Q_{UV,t,90\%}$ (kJ l^{-1})
Chlorine	0.910	1.104	0.62; 2.08	0.15	0.51	0.998	0.419	1.65; 5.48	0.39	1.30
H_2O_2	0.978	0.076	9.12; 30.26	0.94	3.13	0.774	0.061	11.34; 37.70	2.74	9.12
Fenton	0.967	0.151	4.58; 15,23	1.09	3.65	0.987	0.093	7.24; 24.73	1.75	5.96
Control	0.882	0.001	602.6; 2300	144.76	–	0.934	0.001	577.62; 2.300	138.54	–
UV-C	0.988	1.242	0.55; 1.85	0.13	0.44	1.000	1.150	0.60; 2.0	0.14	0.48
SHP	0.999	0.457	1.51; 5.03	0.36	1.58	0.994	0.208	3.32; 11.05	0.79	2.62
SPF	0.999	0.597	1.16; 3.85	0.27	0.922	0.942	0.037	18.48; 62.16	4.43	14.8
SCL	0.976	1.473	0.47; 1.56	0.11	0.37	0.967	1.193	0.57; 1.92	0.13	0.46
sunlight	0.837	0.028	24.47; 82.14	5.87	–	0.881	0.015	45.75; 153.00	10.98	–

bacteria are more resistant than Gram-negative (*E. coli*) bacteria. Accordingly, different disinfection processes showed an higher removal efficiency of *E. coli* compared to Entero. Fiorentino et al. (2019) and Esteban et al. (2017) showed faster bacterial inactivation kinetics in wild Entero and wild *E. coli* in urban wastewater treated by solar driven AOPs. Specifically, in the work of Fiorentino et al. (2019) wild bacteria were tested in three different secondary treated urban wastewater and, in all cases, Entero were difficult to inactivate. A higher resistance to AOPs of Gram-positive bacteria compared to Gram-negative was observed in previous studies (García-Fernández et al., 2019). The Entero inactivation was slower than *E. coli* in solar water disinfection (Berney et al., 2006; Navntoft et al., 2008). Analogous behavior was observed in the case of chlorination. Azuma and Hayashi (2021) observed that Entero were inactivated more slowly than *E. coli* in a hospital effluent; 10 log units of Entero were inactivated only at high doses of chlorine (from 10 mg l^{-1} and up) and after at least 30 min of contact time, whereas for *E. coli* the inactivation time was faster (10 min) as well as the doses (from 3 mg l^{-1}). Some studies have shown opposite behavior between Gram-positives and Gram-negatives. Indeed, Xiao et al. (2020) recently investigated the bacterial inactivation of *E. coli* and Entero in municipal wastewater and deionized water with an initial bacterial load of approximately 1000 CFU mL^{-1}. In their study, a 75% inactivation of Entero was achieved after only 12 min, while it took approximately 80 min to achieve a similar inactivation of *E. coli* using SO_4^- as a disinfecting agent. Luo et al. (2021) in a recent work investigated the inactivation rate of *E. coli* by peroxymonosulfate, two-dimensional transition metal dichalcogenide, and Fe. 97% inactivation of *E. coli* was achieved in 1 min and 4 log units within 5 min. This system generated SO_4^- and HO• radicals even if the fundamental role was played by SO_4^- radicals. This different behavior has also reported in previous studies where Gram-negative bacteria were more resistant to disinfectants due to their complex outer membrane (Fu et al., 2005; Lan et al., 2007). The higher resistance of Gram-negative bacteria can be attributed to their different chemical composition of the cell walls (Demidova and Hamblin, 2005). Thus, it is possible to assume that the disinfecting agent could play a decisive role in the inactivation of Gram-positive and Gram-negative bacteria. Hydroxyl radicals generated by AOPs and disinfectant agents like chlorine damage the cell wall differently, however, despite a different inactivation mechanism, they both inactivate faster Gram-positives than Gram-negative bacteria.

The inactivation rates in solar driven process were much faster than those in dark conditions (Fig. 3).

The sunlight used alone was, as expected, the process with the slowest inactivation kinetics. Only 1 and 0.5 log units of *E. coli* ($k = 0.028\ min^{-1}$, $t_{1/2} = 24.47$ min) and Entero ($k = 0.015\ min^{-1}$, $t_{1/2} = 45.75$ min) were removed, respectively. The fastest inactivation kinetics were observed for the SCL process; 10 and 15 min were enough for the inactivation of *E. coli* ($k = 1.473\ min^{-1}$, $t_{1/2} = 0.47$ min) and Entero ($k = 1.193\ min^{-1}$, $t_{1/2} = 0.57$ min), respectively. In UV-C treatment, 15 and 20 min, were required for *E. coli* ($k = 1.242\ min^{-1}$, $t_{1/2} = 0.55$ min) and Entero ($k = 1.150\ min^{-1}$, $t_{1/2} = 0.60$ min) inactivation respectively. Promising results were obtained with SPF process, as the total bacterial inactivation was get after 25 and 30 min for *E. coli* ($k = 0.597\ min^{-1}$, $t_{1/2} = 1.16$ min) and Entero ($k = 0.037\ min^{-1}$, $t_{1/2} = 18.48$ min), respectively. Total inactivation of *E. coli* ($k = 0.457\ min^{-1}$, $t_{1/2} = 1.51$ min) by SHP process, was achieved only after 60 min, while it was not observed (1 log units residual concentration) for Entero ($k = 0.208\ min^{-1}$, $t_{1/2} = 3.32$ min) even after 60 min contact time. The results from solar driven AOPs tests confirmed the those achieved by dark processes with *E. coli* inactivated faster than Entero. The inactivation kinetics are summarized in Table 2.

In solar driven processes, the inactivation kinetics undergo an important increase, with a consequent reduction of half-life time compared to dark process. K values for *E. coli* inactivation increased from 1.104 min^{-1} for chlorine to 1.473 min^{-1} for SCL, with a $t_{1/2}$ decrease from 0,62 min to 0.47 min. It also increased from 0.076 min^{-1} for H_2O_2 to 0.457 min^{-1} for SHP, with a $t_{1/2}$ decrease from 9.12 min to 1.51 min. Finally, the k value increased from 0.151 min^{-1} for Fenton to 0.597 min^{-1} for SFP, with a $t_{1/2}$ decrease from 4.58 min to 1.16 min. The same behavior was observed for the inactivation of Entero. The slower inactivation rate of Entero compared to *E. coli* by SPF is consistent with our previous work (Fiorentino et al., 2019) where the bacterial inactivation of indigenous bacteria in a secondary effluent by photo Fenton like using IDS-Cu showed a 3.5 log units inactivation of *E. coli* after 10 min, resulting very similar to the bacterial inactivation time found in this work. Noteworthy, in that case the concentrations of the photocatalytic reagents were 4 times lower, as well as the initial bacterial load. Several works have investigated bacterial inactivation by solar photo Fenton process (H_2O_2/Fe^{2+}/sunlight) in secondary treated wastewater (Ferro et al., 2015; Rodríguez-Chueca et al., 2014; Villegas- Guzman et al., 2017). Rodríguez-Chueca et al. (2014) detected a complete *E. coli* inactivation (6 log units) after 120 and 180 min for indigenous *E. coli* and Entero respectively at 10/50 mg l^{-1} of H_2O_2/Fe^{2+} at pH 5. In Ferro et al. (2015), a complete *E. coli* (6 log units) inactivation was achieved after 180 min at pH 8.5, at 0.090/0.294 mM of H_2O_2/Fe^{2+}, respectively. In our work, the results of SCL process are promising: a reduction of contact time higher than 50%, compared to treatment with chlorine alone, was observed. To the best of authors' knowledge, SCL process has not been previously investigated in the inactivation of *E. coli* and Entero, however it was found to be effective in the inactivation of *Cryptosporidium* oocyst (Zhou et al., 2014) and *Bacillus subtilis* endospores (Forsyth et al., 2013). Wang et al. (2012) investigated sequential chlorine/UV-C process, and a total inactivation of heterotrophic bacteria was achieved with 1.6 mg l^{-1} unlike of the 5.5 mg l^{-1} necessary for chlorine alone. The bacterial inactivation in gray water by sunlight through a compound parabolic collector reactor was investigated by Strauss et al. (2018) and 8 h treatment was necessary to achive a total inactivation of *E. coli* (1 log units). H_2O_2/IDS-Cu/sunlight and chlorine sunlight did show an excellent inactivation efficiency, even if the mechanisms of cell damage are different. In SPF by IDS-Cu the damage can be produced by different actions. In particular, different mechanisms take place simultaneously during SPF

at neutral Ph, as described by García-Fernández et al. (2019) and de la Obra Jiménez et al. (2020). The mainly mechanisms involved for bacterial inactivation include: (i) the generation of external hydroxyl radicals in Fenton and photo-Fenton reactions which attack the cellular membrane; (ii) DNA mutation due to intracellular HO• formation because of UVA radiation and the action of other Reactive Oxygen Species (ROS) generated and accumulated during solar exposure; and (iii) the diffusion of H_2O_2 into the cell. On the contrary, different oxidizing species are generated by sunlight/chlorine as described by Yang et al. (2016). In particular, the (i) HO•, (ii) ozone production from atomic oxygen ($O(^3P)$) (according to Eqs. (7) and (8)) (40.6%), and (iii) HOCl (the latter in a very mild way) are they are responsible for the degradation process.

$$OCl^- + h\nu \rightarrow O(^3P) + Cl^- \quad (7)$$

$$O(^3P) + O_2 \rightarrow O_3 \quad (8)$$

To authors' knowledge, there are no disinfection investigations by sunlight/chlorine, from the experience of this work, it emerged that the radical species generated by this process are highly efficient in bacterial inactivation and competitive with SFP.

3.3. Bacterial regrowth after disinfection treatment and subsequent storage

The regrowth of Entero and *E. coli* after an apparent complete bacteria inactivation was investigated at different post-treatment contact times (6, 12, 24 and 48 h) for all disinfection processes (Fig. 4). The investigation of possible bacterial regrowth is really important, in particular when disinfected water are stored after treatment, before reuse

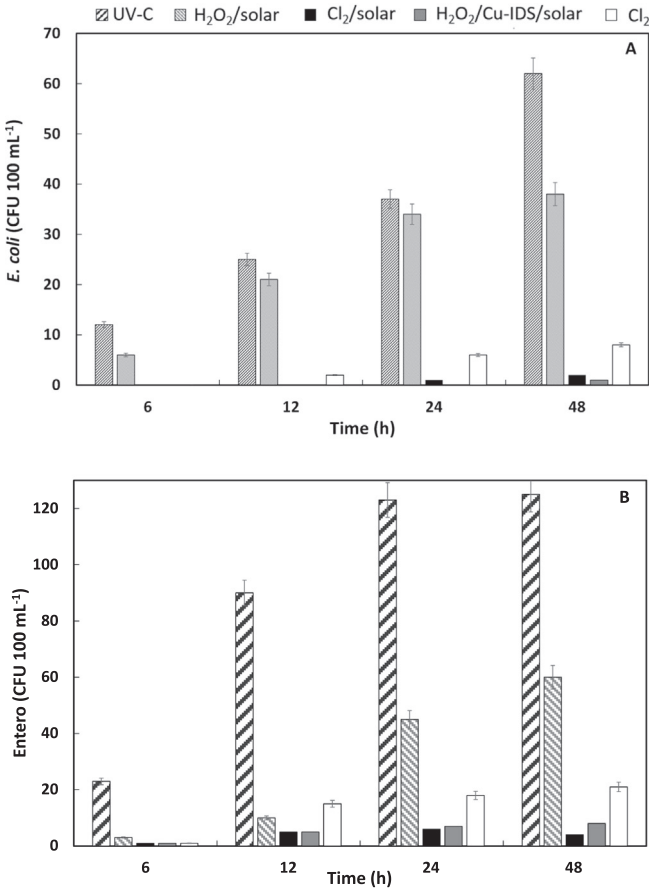

Fig. 4. Regrowth of *E. coli* (A) and Entero (b) at 6, 12, 24 and 48 h contact time after complete inactivation by the investigated disinfection processes.

for crops irrigation (European Commission. Directorate General for Health and Food Safety, 2017). Bacterial cells only injured by disinfection process cannot regrowth on standard cultivation media which may overestimate disinfection process efficiency (Rizzo et al., 2004). Therefore, if the post-treatment environmental conditions allow it, bacterial cells can repair the injury and contribute to bacterial regrowth.

SCL and SPF processes possibly resulted in a higher damage to bacterial cells because the lower post-treatment regrowth was observed (<10 CFU ml^{-1}, limit set by EU water reuse regulation for *E. coli*, for all incubation times) (Fig. 4). SHP process showed a slower bacterial inactivation than SPF which finally affected subsequent regrowth. Under all the disinfection processes and conditions tested, Entero showed a higher regrowth potential than *E. coli* with a slight regrowth already detectable after 6 h post-treament. Possibly, the cell structure of Entero, and thus of Gram-positives, not only protects bacterial cells to some extent from the inactivation, but also results in an easier regrowth of damaged bacterial cells. Finally, the UV-C process was the less effective process in contrasting bacterial regrowth; indeed after 24 h, 40 and 120 CFU ml^{-1} of *E. coli* and Entero were detected. The extremely limited regrowth achieved by the SPF process is also confirmed by previous works (Giannakis et al., 2015, 2016). The application of solar photo-Fenton combines the simultaneous exposure to solar UV and the Fenton reagents, and this can limit the potential for recovery of microorganisms. No study has analyzed bacterial regrowth after disinfection by SCL so far, to authors' knowledge. It is very likely that radicals formation following chlorine photolysis by sunlight created unfavorable conditions for regrowth, as the regrowth was higher than chlorination alone. Another aspect to consider in regrowth is the low organic content of RHRW compared to wastewater. The higher organic loading of wastewater can promote a more easy bacteria cells aggregation and create substrates where they can be reactivated after the disinfection process (Giannakis et al., 2016). In this case, the low organic matter loading of RHRW compared to urban wastewater has contributed to the lower oxidant demand of the aqueous matrix, making the relatively low doses of reagents sufficient to limit bacterial regrowth after 48 h in SPF and SCL processes. *E. coli* bacterial regrowth after chlorination and sunlight/H_2O_2 processes, respectively, was investigated by Fiorentino et al. (2015). Unlike of the higher inactivation efficiency of the chlorine process, the damage to bacterial cells from sunlight/H_2O_2 process resulted in a comparatively lower regrowth of bacterial cells.

4. Conclusions

The SPF process, using IDS-Cu complex as catalyst under sunlight and investigated for the first time as disinfection process, provided really encouraging results in terms of bacterial inactivation and regrowth in RHRW. SCL was the most effective disinfection process ($k = 1.473\ min^{-1}$, $t_{1/2} = 0.47$ min for *E. coli* and $k = 1.193\ min^{-1}$, $t_{1/2} = 0.57$ min for Entero). Although UV-C radiation (0–1.3×10^4 μW s cm^{-2} dose range) was the second faster disinfection process ($k = 1.242\ min^{-1}$ and $t_{1/2} = 0.55$ min for *E. coli*), it was the less effective process in controlling bacterial regrowth, making the effluent not suitable for crop irrigation already after 6 h post-treatment storage, if the limit for *E. coli* (10 UFC 100 ml^{-1}) set by EU for wastewater reuse is taken as reference value. SPF and SCL are interesting options for RHRW disinfection, especial in case of effluent use for crop irrigation, however further investigation is necessary before to make a conclusion on the most effective and sustainable process. In particular, the formation of possible toxic oxidation intermediates should be taken into account through toxicity tests as well as the formation of chlorination by products should be evaluated in SCL processes, although a lower formation is expected compared to chlorination processes (Cerreta et al., 2020). Finally, the implementation of life cycle assessment, taking all these end points into account, would be an useful tool to make a decision about the best treatment option.

CRediT authorship contribution statement

A. Fiorentino: Conceptualization, Investigation, Formal analysis, Writing – original draft. **G. Lofrano:** Conceptualization, Supervision, Writing – original draft. **R. Cucciniello:** Validation, Writing – original draft. **M. Carotenuto:** Conceptualization, Validation. **O. Motta:** Conceptualization, Resources, Funding acquisition. **A. Proto:** Conceptualization, Supervision, Resources, Funding acquisition. **L. Rizzo:** Conceptualization, Supervision, Writing – review & editing.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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