

Solar disinfection of wild *Salmonella sp.* in natural water with a 18 L CPC photoreactor: Detrimental effect of non-sterile storage of treated water

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Abstract

For the first time solar disinfection of liters of water containing wild *Salmonella sp.* and total coliforms was carried out in a compound parabolic collector (CPC) photoreactor at temperatures of almost 50 °C. Using surface water with high turbidity, this treatment was efficient in completely inactivating *Salmonella sp.* without regrowth during the subsequent 72 h of dark sterile storage. However if the solar treated water is poured in a non-sterile container, bacteria regrowth occurs even if 10 mg L⁻¹ of H₂O₂ is added before the storage. On the other hand, 30 mg L⁻¹ of H₂O₂ added when the irradiation started was completely depleted within 2 h and did not prevent bacterial regrowth during post-irradiation storage in non-sterile containers, demonstrating that storage of large volumes of water treated by solar irradiation was not optimal. Finally, total coliforms (*Escherichia coli* included) showed a far higher sensitivity than *Salmonella sp.* and demonstrated to be an inappropriate indicator for monitoring bacterial contamination in water during solar disinfection processes. © 2011 Elsevier Ltd. All rights reserved.

Keywords: Solar water disinfection; SODIS; CPC photoreactor; Post-irradiation events; *Salmonella sp.* inactivation

1. Introduction

Many Sahelian countries undergo drinking water supply problems. In Burkina Faso, surface water collection was developed through dam constructions. However, surface waters are often affected by human activities, which imply hazardous chemicals and especially bacteriological pollution. The socio-economical context, mainly in isolated and rural populations (Boyle et al., 2008), considerably reduces the applicable techniques for water disinfection. In addition, chlorination, which is a largely used technique

for surface water disinfection, is limited by the formation of toxic by-products such as trihalometanes (THMs), resulting from the reaction of chlorine with natural organic matters (NOM) often present in surface waters (Bond et al., 2009; Moncayo-Lasso et al., 2009; Mosteo et al., 2009).

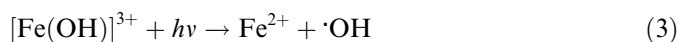
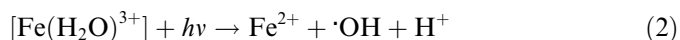
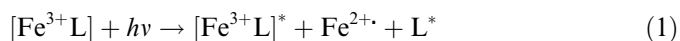
In the sunny regions of Burkina Faso, it is of interest to develop low cost disinfection methods such as SODIS. Solar water disinfection in bottles, known as the SODIS process, is a simple and widely used household technique constituting a small-scale way to improve water quality for drinking purposes at point-of-use (Du Preez et al., 2010; Wegelin et al., 1994). SODIS treatment for a whole household unit requires a large configuration (in terms of

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bottles exposed and stored at the same time) and around 6 h of solar exposure, which can increase during cloudy days to up to 48 h, followed by storage to cool the water (Oates et al., 2003). Bacterial inactivation by SODIS treatment is the consequence of two synergistic factors: (i) the effect of UV-A (between 320 and 400 nm) and the visible irradiation between 400 and 490 nm, and (ii) a temperature increase which must reach at least 45 °C (McGuigan et al., 1998; Sommer et al., 1997; Wegelin et al., 1994). SODIS treatment for water disinfection has often been evaluated through the monitoring of *Escherichia coli* inactivation as the main bacterial indicator. However, Berney et al. (2006) have found that this microorganism is one of the most sensitive to the effect of solar irradiation and temperature with regard to other waterborne human pathogens. For this reason, it does not represent a suitable indicator of SODIS performance for drinking water production. In contrast, *Salmonella typhimurium* an extremely pathogenic microorganism, seems to be more resistant to SODIS treatment, making it a far more suitable indicator (Berney et al., 2006; Smith et al., 2000; Winfield and Groisman, 2003; Rincón and Pulgarin, 2003).

In natural surface waters, humic substances and organic chromophores (which are part of NOM), act as photosensitizers. Under UV–Vis irradiation, these substances induce O₂ reduction leading to reactive oxygen species (ROS) such as singlet oxygen (¹O₂), superoxide (HO₂[•]/O₂^{•-}), and [•]OH radicals very toxic to cells (Canonica, 2007; Moncayo-Lasso et al., 2008a; Paul et al., 2004). Hence, dissolved oxygen concentration in water is considered as a directly influencing parameter of the bactericidal action of SODIS treatment (Curtis et al., 1992; Gourmelon et al., 1994; Reed, 1997). As agitation can promote the release of dissolved oxygen, potential SODIS reactor configurations have to be evaluated individually with regard to this parameter (Kehoe et al., 2001).

Next to this, sunlight can also induce other ROS-producing reactions in natural waters containing transition metals such as iron, which is the case in Sahelian regions, where water flows on ferruginous substrates (Ben Yahmed, 2005). At a natural pH of 7.5, iron species in aqueous solution are principally present as organo- (and aqua-) complexes of ferric ions (Gallard et al., 1999; Moncayo-Lasso et al., 2008b; Pignatello et al., 2006). Under visible light, the photoreduction of dissolved ferric ions via ligand-to-metal-transfer (LMCT) reaction also leads to the production of [•]OH radicals (Goslan et al., 2006; Lee and Yoon, 2004; Malato et al., 2009; Vermilyea and Voelker, 2009):



Recently, some of us have reported that in synthetic or real waters at near neutral or neutral pH, photo-sensible iron species could directly interact with cell membranes (complexation with proteins and other membrane compo-

nents) avoiding its precipitation and playing an important role in photo-Fenton disinfection processes (Sciacca et al., 2010; Spuhler et al., 2010).

Despite its simplicity, SODIS point-of-use treatment has not been fully explored to disinfect larger volumes of drinking water. PET bottle exposition on a corrugated iron sheet implies treatment in series with different sets of bottles in order to obtain enough water for a household family. The SODIS principle has been improved through changes in its configuration, which could also allow the disinfection of larger water volumes than conventional PET bottles (Kehoe et al., 2001; Navntoft et al., 2008). Among the possibilities evaluated, reflection of solar irradiation around the exposed reactor pointed out the advantages of temperature increase on enhancement of bacterial inactivation (Martín Domínguez et al., 2005; Rijal and Fujioka, 2003). In this context, using of compound parabolic collector (CPC) photoreactor showed an evident capacity to enhance solar disinfection by photocatalysis reactions while non-TiO₂ photo-assisted treatment is not efficient (Fernández et al., 2005; Rincón and Pulgarin, 2005; Rincón and Pulgarin, 2007a,b).

Ubomba-Jaswa et al. (2010) have recently demonstrated that treating large water volumes (25 L) by CPC photoreactors under solar irradiation could efficiently inactivate *E. coli* K-12 cells without regrowth within 48 and 72 h following the solar disinfection. However, solar disinfection in a CPC reactor has still not been evaluated in areas with a constant solar irradiation and in natural waters containing wild bacteria strains. This study aims to evaluate the disinfection efficiency of the solar irradiation process (reproducing field conditions) in a CPC solar photoreactor with natural surface water sampled in urban dams containing wild *Salmonella sp.* of Ouagadougou, Burkina Faso. Experimentation was carried out during April–May 2009 when air temperature was of about 40 °C (see Fig. 1).

2. Experimental section

2.1. Reactors

2.1.1. Preliminary experiments at laboratory scale – solar simulator

Pirex-glass bottles of 50 mL were used as batch reactor and exposed under simulated solar light. A Hanau Suntest solar simulator (placed in laboratory facilities of the Institut International d'Ingénierie de l'Eau et l'Environnement (2iE) in Ouagadougou-Burkina Faso) is equipped with a Xenon lamp, which has a spectral distribution of about 0.5% of the emitted photons at wavelengths shorter than 300 nm (UV-C range) and about 7% between 300 and 400 nm (UV-B, -A range). Simulated solar spectrum between 400 and 800 nm follows solar spectrum. Experiments were carried out using a solar simulated irradiation with a light global intensity of about 660 W m⁻² (32 W m⁻² on the UV-A range).

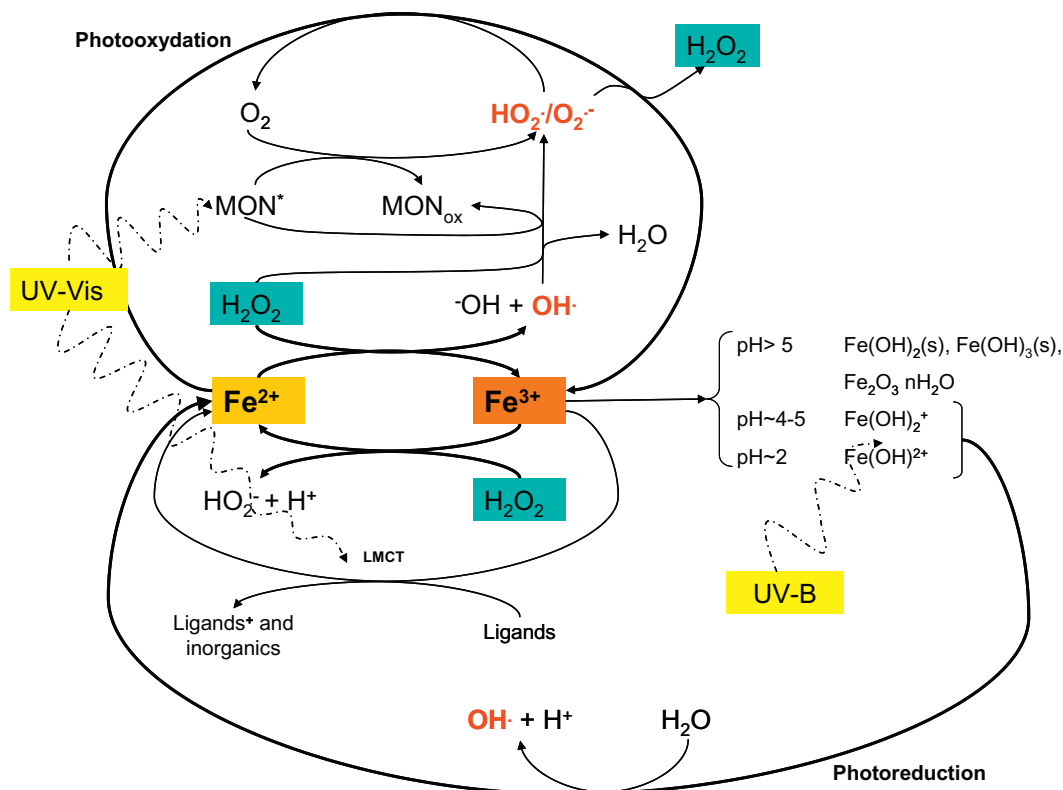


Fig. 1. Potential reaction pathways of the helio-photo-Fenton process (Moncayo-Iasso et al., 2009; Rincón and Pulgarin, 2007a; Pignatello et al., 2006; Bandara et al., 1996; Sun and Pignatello, 1993).

2.1.2. Experiment at field scale – CPC solar photoreactor

Solar pilot plant SOLARDETOX[®] ACADUS 2003 distributed by Ecosystem S.A. (Barcelona, Spain), is composed of 16 borosilicates cylindrical glass tubes of 32 mm diameter, 1.5 m length and 1.4 mm of width, through which water is circulating and exposed to solar irradiation (Fig. 2b). Tubes are disposed in such a way that the cylindrical-parabolic mirrors distribute UV irradiation equally around the whole tube circumference (Fig. 2c). This configuration implies no light concentration, but allows working with diffuse light. The CPC module of exposition is characterized with a useful collector surface of 2.12 m², a total surface of 2.54 m², a photoreactor active volume of 15.1 L, a total volume of 16.07 L and a working volume between 18 L and 50 L. A polypropylene stirred tank of 50 L is connected in series with the CPC module and constitutes a re-circulating tank. Hence, the pilot plant behaves as a plug-flow reactor where water is circulating using a centrifugal pump with a flow of 24.2 L min⁻¹. The CPC photoreactor was positioned with 10° of inclination corresponding to the approximate local latitude of Ouagadougou (Burkina Faso). Similar CPC solar photoreactors have been previously described elsewhere (Malato et al., 2009; Sarria et al., 2003; Sichel et al., 2007; Rincón and Pulgarin, 2007b).

Solar ultraviolet radiation is determined during the experiments by means of a UV-A radiometer ACADUS

85 UV (Fig. 2a) fixed on the CPC photoreactor at the same inclination (10°). Solar intensity per square meter (W m⁻²) is consecutively monitored between 300 and 400 nm on the spectral regions.

2.2. Analytical methods

Temperature and pH were measured with a *Universal pocket meter* WTW 340i equipped with a WTW SenTix 41-3 prob sonde. Natural pH was near neutral and was not modified during the experiments. Turbidity was determined with the nephelometric method described in APHA Standards Methodology (APHA, 2001). Total iron was evaluated with the HACH FerroVer method using a HACH 2010 spectrophotometer and UV absorbance at 254 nm with a Biomate 3 model. Total organic compounds (TOC) were monitored using a Shimadzu 500. Before measuring, each sample was stored at 4 °C after addition of sodium bisulphite to eliminate the H₂O₂ and stop any Fenton and photo-Fenton reactions. Experimental configuration at laboratory scale permitted to monitor the bacterial inactivation corresponding to SODIS process and temperature increase twice (in two different glass bottles). Minimum and maximum values of bacterial concentration are the average of three determinations. At field scale, each sample was done only once with a sterile flask

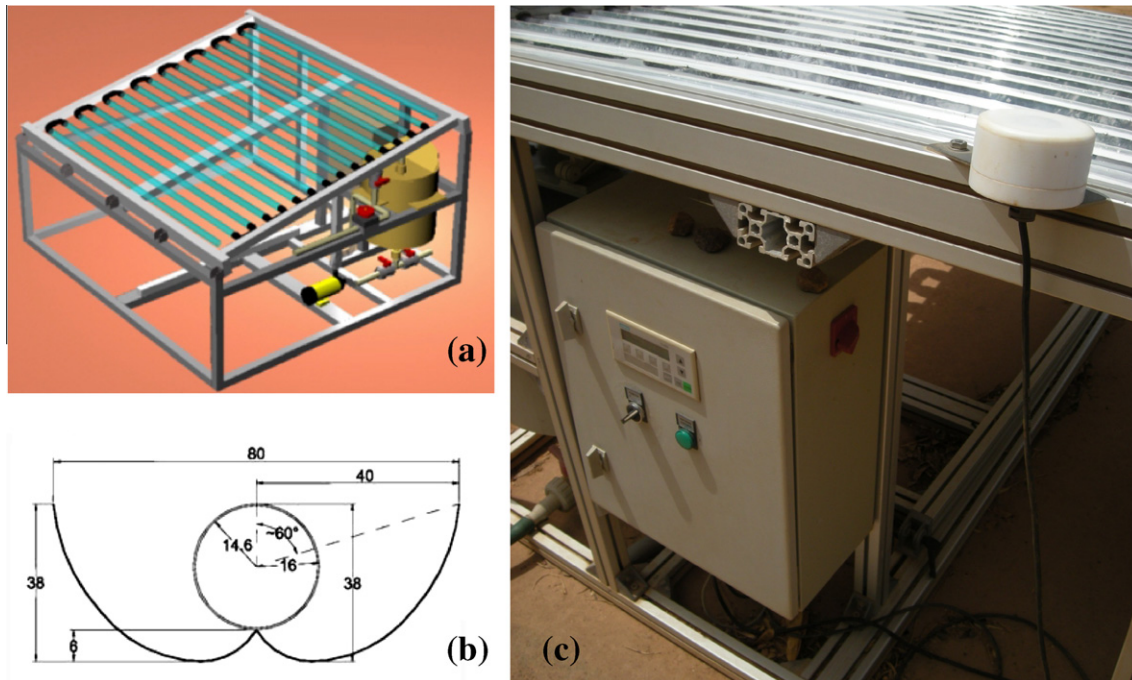


Fig. 2. (a) Schematic view of the photoreactor used at 2IE (Ouagadougou, Burkina Faso), Compound Parabolic Collector (CPC); (b) reflective surface describing an involute around a cylindrical tube on the CPC, the units of measurements are mm, and (c) view of the solar UV-A radiometer.

of 15 mL and settled just before being plated to monitor bacterial inactivation.

2.3. Water sources

Dam surface water (urban dam no. 1, Ouagadougou) was sampled 1 day before the irradiation experiment and stored at 4 °C. Composite samples from three different places around dam no. 1 were used to carry out laboratory and field scale experiments in order to have similar physico-chemical and bacteriological characteristics. Nevertheless, a distinction between well water of dam no. 1 (dry period) and backwater of dam no. 1 (beginning of rainy season) is made because of noticeable differences in water quality. Some physico-chemical and bacteriological characteristics of the treated water are given in Table 1.

2.4. Water illumination procedures and bacterial sampling

2.4.1. Solar disinfection experiments carried out at laboratory scale

At laboratory scale under simulated solar light (Hanau-Suntest), nine glass bottles of about 50 mL were used in parallel and filled with natural surface water. The experimental configuration was composed of three irradiated glass bottles, three irradiated glass bottles protected by an aluminium sheet and three reference solutions in the darkness. One bottle of each different configuration was used only to monitor temperature evolution, whereas the two others were kept to monitor only bacterial inactivation. Just before starting illumination, each bottle was sam-

pled to determine the initial bacterial count. The solutions were irradiated for 4 h, at ambient laboratory temperature (30.9 °C) and up to 49.9 °C for solar disinfection experiments, 49.4 °C for the temperature effect solutions (wrapped with aluminium foil sheet) and 40.6 °C for the reference dark solutions. Samples were taken every 15 min until 30 min of simulated irradiation, every 30 min until 2 h and every hour until the end of the experiment (4 h). Serial dilutions were performed in deionized water. Taken samples were immediately spotted onto Chromocult® Coliform agar (Merck) and spread using standard techniques. This selective agar allows the identification of different kinds of bacteria colonies through different colours: total coliforms salmon to red color and *Salmonella* colourless. Colonies were counted after 24 h at 37 °C in the dark. Bacterial trends were made with two main populations: Total coliforms and *Salmonella sp.* Then, the bottles were stored in the dark at ambient temperature and sampled at 24, 48, and 72 h after the end of the experiment to monitor post-irradiation events.

2.4.2. Solar disinfection experiments ran at field scale

At field scale in the CPC solar photoreactor, a 18 L sample was taken out the fridge 2 h before experiment and stored at ambient temperature, which started at 10:00 a.m. for 4 h. During the experiments, water temperature often reached values above 50 °C. The same approach as at laboratory scale was used to follow bacterial inactivation. UV-A solar radiation intensity (W m^{-2}) during the experiments was monitored with an ACADUS 85 UV radiometer. The average and maximum UV-A intensity are

Table 1

Physico-chemical and bacteriological parameters of the urban dam no. 1 in Ouagadougou. Two types of water were sampled; surface water from wells in dam basin and surface water from backwater (treated only with CPC solar photoreactor).

Parameter	Suntest and CPC experiments + 30 mg L ⁻¹ of H ₂ O ₂ . Dam no. 1 surface water from wells (Figs. 3a, b and 5)	CPC experiments. Dam no. 1 surface water from wells (Fig. 4a)	CPC experiments. Dam no. 1 surface water from backwater (Fig. 4b)
<i>T</i> (°C)	28.9	28.8	29.2
pH	6.9	6.9	8.0
Turbidity (NTU)	78	76	>1000
Iron total (mg L ⁻¹)	1.3	1.2	17.0
TOC (mg L ⁻¹)	9.3	9.2	63.9
Total coliforms (CFU mL ⁻¹)	3.9 × 10 ³	5.7 × 10 ³	10 × 10 ³
<i>Salmonella sp.</i> (CFU mL ⁻¹)	1.4 × 10 ³	1.8 × 10 ³	2.0 × 10 ³

reported. After the solar disinfection treatment, water was stored in the dark at ambient temperature in sterile (washed with diluted hypochlorite solutions) and non-sterile containers (washed with crude water to simulate the worse case) and sampled 24, 48 and 72 h.

3. Results and discussion

3.1. Total coliforms and *Salmonella sp.* inactivation by temperature increasing and simulated solar irradiation exposure at laboratory scale (50 mL)

Two experiments were carried out in parallel with surface water of urban dam no. 1 (table 1) at natural neutral pH (6.9). Fig. 3a and b shows that internal temperature under simulated solar irradiation (Suntest) exceeded 45 °C after 30 min of exposition and reached a maximum of 49.4 °C, whereas bottles exposed to the light but protected with aluminium foil sheets reached 49.9 °C and only 41 °C in the dark (non-irradiated bottles). With only heating produced in samples covered with aluminium foil sheets, total coliforms (*E. coli* included) were completely inactivated without further regrowth. The coupled effect of simulated solar irradiation and temperature in illuminated samples enhanced the inactivation rate of total coliforms. In contrast, *Salmonella sp.* concentration underwent a 3 logs reduction by temperature rise under aluminium foil in 4 h, whereas it was totally inactivated in 1 h 30 min under the simultaneous action of temperature and solar irradiation in illuminated samples. However, in both cases bacterial recovery was observed during subsequent dark storage at ambient temperature of 35 °C.

Total coliforms are thus much more sensitive than *Salmonella sp.* to both temperature rise and solar irradiation. This confirms that inactivation mechanisms can be different according to the enteric pathogen considered as proposed by Berney et al. (2006). In previous studies, *Salmonella sp.* inactivation under sunlight irradiation demonstrated a noticeable resistance compared to *E. coli* (Evison, 1988). Especially in the SODIS disinfection context, a pathogen bacterium such as *Salmonella sp.* should often be considered as an indicator of the disinfection effectiveness. We also observed this effect whilst carrying out

PET bottles experiments in the same geographical context (Sciacca et al., 2010).

3.2. Total coliforms and *Salmonella sp.* inactivation in an 18 L CPC solar photoreactor at field scale

Results with the CPC solar reactor containing well water from urban dam no. 1 (Table 1) under solar UV-irradiation with average and maximum intensities of respectively 31.7 W m⁻² and 35 W m⁻² are shown in Fig. 4a. Both *E. coli* and *Salmonella sp.* were completely inactivated after 1 h 30 min and 3 h respectively, without any observed regrowth for 72 h of subsequent dark sterile storage. Temperatures exceeded 45 °C after 30 min exposure and reached a maximum of 49.2 °C.

Fig. 4b shows the results obtained with backwater from urban dam no. 1 which is much more turbid and charged with NOM (Table 1), for average and maximum UV-intensities of respectively 34 W m⁻² and 44 W m⁻². Complete inactivation was reached after 1 h 30 min of illumination, without any observed regrowth for 72 h of subsequent dark sterile storage. Temperature exceeded 45 °C just after 30 min of exposition and reached a maximum of 54.2 °C. These solar disinfection results with CPC could open the possibility for treating larger water volumes than with a simple SODIS in PET bottles. CPC is not efficient for bacterial inactivation when high temperatures are not reached, that is probably due to its plug-flow configuration which requires dark spaces like the intermediate tank and the unexposed circulating system that moreover could favour bacteria dark repairing and has a detrimental effect on solar disinfection. In this context, Ubomba-Jaswa et al. (2009) maintained that a UV dose received in an interrupted way has much less inactivation potential when confronted with a non-recirculating reactor (uninterrupted exposition). The significant rise of temperature in backwaters is probably helped by the presence of high organic and mineral suspended matter (76–1000 NTU) that act as a thermic carrier, compensating its negative impact on light penetration.

Note that using data obtained with the solar simulator to predict results obtained with the CPC reactor is not appropriate, especially because pumping shear forces

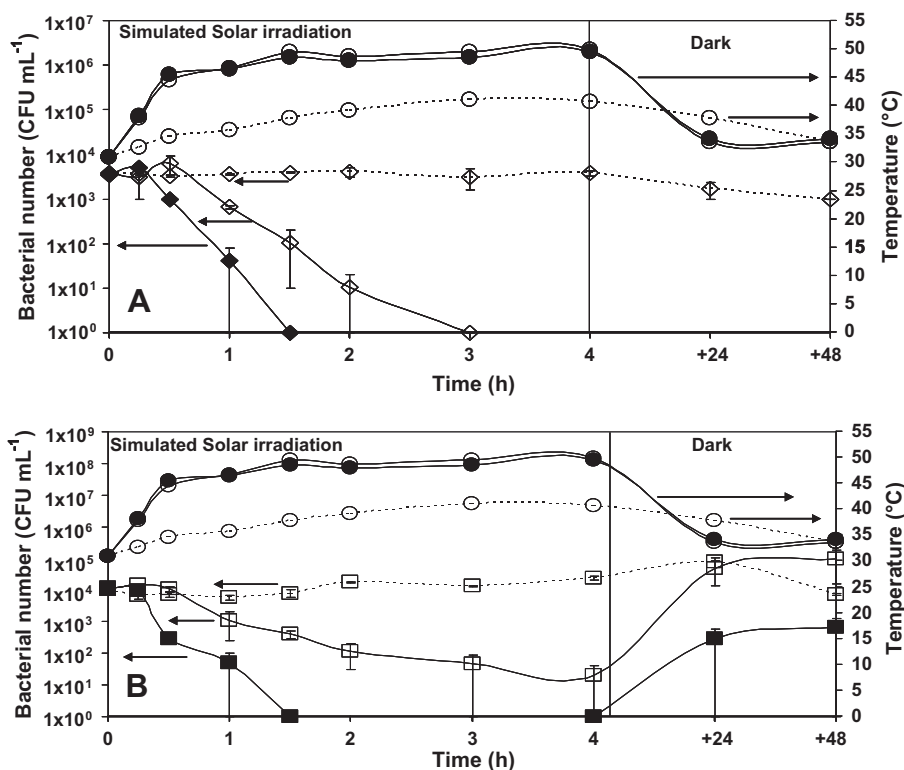
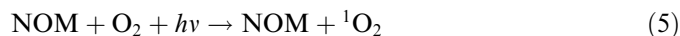
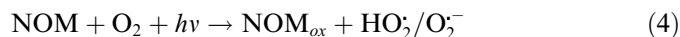


Fig. 3. Bacterial inactivation in water of dam 1 (see Table 1) by simulated solar light at 32 W m^{-2} of UV. Temperature ($^{\circ}\text{C}$) in glass bottles ($-\bullet-$), in glass bottles under alu. sheet ($-\circ-$) and in dark bottles out of solar simulator ($-\diamond-$). (A) Total coliforms. Simulated solar treatment ($-\blacklozenge-$), simulated solar light covered with aluminium sheet ($-\square-$) and in the dark ($-\diamond-$). (B) *Salmonella sp.* simulated solar treatment ($-\blacksquare-$), simulated solar light covered with aluminium sheet ($-\square-$) and in the dark ($-\square-$).

produced by recirculation of water in CPC might weak the bacterial biomass and affect the inactivation process. Moreover, recirculation of water in CPC modifies oxygen concentration. Kehoe et al. (2001) tested the effect of stirring on *E. coli* inactivation during solar treatment in small volumes (20 mL) and observed a longer inactivation time explained by the decrease in oxygen content. On the other hand Ubomba-Jaswa et al. (2009) also observed a longer inactivation time for *E. coli* with the increase of CPC recirculation flow rate from 0, 2 to 10 L min^{-1} . However, our CPC experiments were carried out using a much stronger flow rate of 24.5 L min^{-1} . A similar flow rate (22.5 L min^{-1}), tested by Fernández et al. (2005), exhibited a noticeable enhancement of bacterial inactivation during both dark and solar exposed experiments with respect to no flow rate at all. Enhanced bacterial inactivation under high recirculation rates could be explained principally by mechanical shear stress. Moreover high oxygen concentration maintained by the hydraulic recirculation in solar CPC experiments favours the ROS production from illuminated photosensitizers, such as natural organic matter (NOM) and Fe^{3+} naturally present in surface waters.

Indeed, humic substances and organic chromophores (contained in NOM) under UV–Vis irradiation and in presence of O_2 can lead to ROS production, such as singlet oxygen ($^1\text{O}_2$), superoxide ($\text{HO}_2^{\cdot}/\text{O}_2^{\cdot-}$) (Eqs. (4) and (5)), and $\cdot\text{OH}$ radicals, which are toxic to bacterial cells (Canonica, 2007; Moncayo-Lasso et al., 2008a; Paul et al., 2004).



ROS production can also be mediated by the combined action of mineral particles such as clay, and photoactive organic particles. Indeed, under solar irradiation, the aggregation of organic photosensitizers on mineral particles shows the ability to produce singlet oxygen, which enhances the antimicrobial effect (Bujdák et al., 2009).

Moreover, at neutral pH, iron species are often under ferric-complex forms like $\text{Fe}^{\text{(III)}}\text{-humate}$. Thanks to ligand-to-metal charge-transfer (LMCT), an intermolecular photoredox process allows the dissociation of these complexes under sunlight and the production of oxidative organic radicals and $\cdot\text{OH}$ radicals (Sarria et al., 2003; Sun and Pignatello, 1993).

3.3. Detrimental effect of H_2O_2 addition on total coliforms and *Salmonella sp.* inactivation in 18 L CPC photoreactor at field scale

The addition of 30 mg L^{-1} of H_2O_2 in CPC experiments (Fig. 5) with well water from urban dam no. 1 (Table 1) under solar irradiation of 4 h (average and maximum UV-intensities of respectively 22.1 W m^{-2} and 30 W m^{-2}) show that both total coliforms and *Salmonella sp.* were inactivated completely after 3 h. Temperature reached a maximum of $43.5 \text{ }^{\circ}\text{C}$ and hydrogen peroxide was exhausted

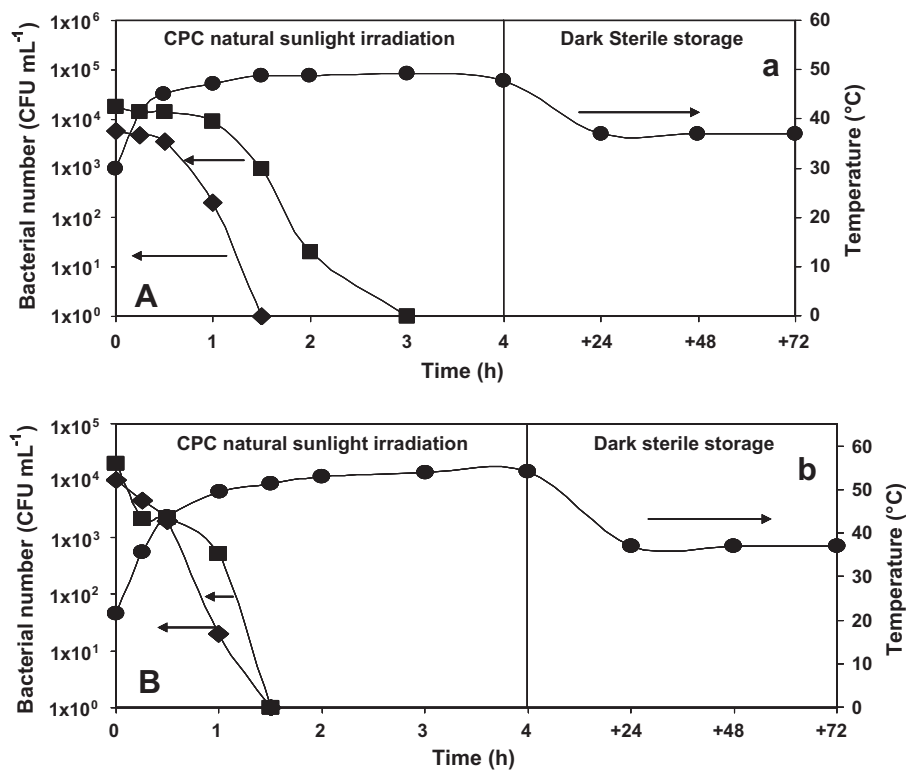


Fig. 4. Bacterial inactivation by CPC solar irradiation (18 L). (A) Well water of urban dam no. 1. Total coliforms (\blacklozenge) and *Salmonella sp.* (\blacksquare). Temperature ($^{\circ}\text{C}$) (\bullet). pH = 6.9. Average UV intensity: 31.7 W m^{-2} . (B) Backwater of urban dam no. 1. Total coliforms (\blacklozenge) and *Salmonella sp.* (\blacksquare). Temperature ($^{\circ}\text{C}$) (\bullet). pH = 8. Average UV intensity 34.8 W m^{-2} .

after less than 2 h of solar exposition. However, during subsequent dark sterile storage, a sharp regrowth of *Salmonella sp.* was observed after 24 h, while total coliforms regrowth was observed after 48 h.

Compared with a simple solar CPC treatment (Fig. 4a), the water samples in which 30 mg L^{-1} of H_2O_2 were added before starting irradiation needed more time to inactivate the bacterial charge and could not avoid bacterial recovery during the subsequent dark sterile storage. Hydrogen peroxide was hence not capable to compensate the limited temperature increase (only $43.5 \text{ }^{\circ}\text{C}$). Indeed, hydrogen peroxide was completely consumed in less than 2 h, whereas microorganisms were under detection limit after only 3 h, which indicated that the oxidant was surely consumed reacting with others organic compounds, typically NOM. The presence of natural iron in water as ferric-complexes could, as already explained in the previous section, catalyse the production of $\cdot\text{OH}$ radicals, which are known to attack almost every organic compounds. This photocatalytic process has shown a noticeable capacity to reduce NOM in natural water (Fukushima and Tatsumi, 2001; Goslan et al., 2006; Moncayo-Lasso et al., 2009; Murray and Parsons, 2004; Pignatello et al., 2006). In addition, humic substances and organic chromophores, which are usually part of NOM, are also considered as photosensitizers. They can produce ROS or directly oxidize other organic compounds in their excited states. Therefore, through solar irradiation or the attack of $\cdot\text{OH}$ radicals, they are easily

degraded and transformed into optimal bacteria substrate like carbonyl groups or amino-acids (Canonica, 2007; Paul et al., 2004). The increase of organic matter bio-available for microorganisms and the absence of residual hydrogen peroxide at the end of the CPC solar exposition are thus probably the main reasons of the regrowth observed.

3.4. Post-irradiation events in CPC photoreactor: storage of treated water

Surface well water from dam no. 1 previously exposed to solar irradiation in the CPC photoreactor as shown in Fig. 4a, was stored according to real and practical non-sterile conditions. Indeed, solar treatment of large volumes of water as proposed by a CPC reactor implies successive distribution in potentially contaminated containers. This is why different configurations of storage have been tested including sterile conditions and non-sterile conditions, with the addition of 2.5 and 10 mg L^{-1} of H_2O_2 monitoring the regrowth for 72 h. Non-sterile conditions are defined as containers rinsed with crude untreated surface water.

After CPC solar treatment of surface well water from dam no. 1, sterile conditions of storage have not shown any bacterial recovery for 72 h (Fig. 4a). In contrast, non-sterile conditions of storage undergo unilateral regrowth of *Salmonella sp.* despite of H_2O_2 addition (Fig. 6). The addition of 2 and 5 mg L^{-1} of H_2O_2 could not avoid the regrowth after 24 h of storage. In contrast,

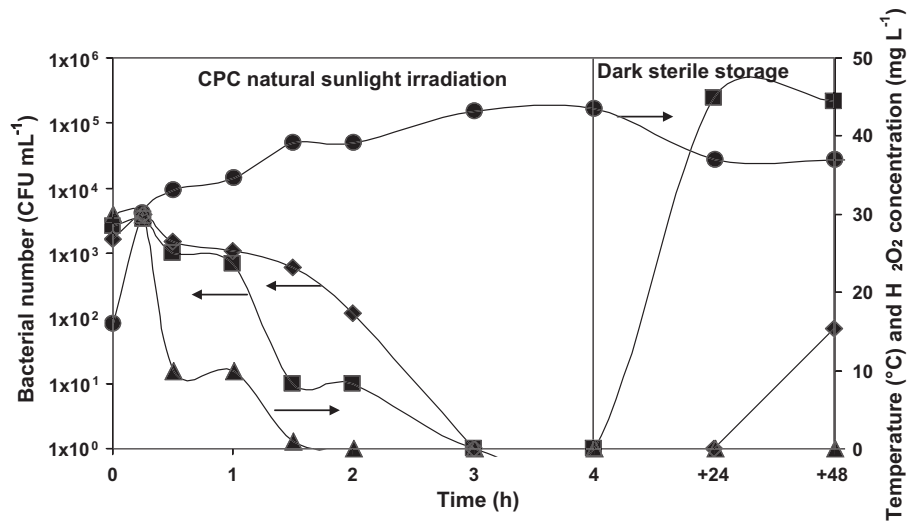


Fig. 5. Bacterial inactivation by CPC solar irradiation (18 L) and addition of 30 mg L⁻¹ of H₂O₂. Average UV intensity 22.1 W m⁻². Well water of urban dam no. 1. Total coliforms (◆) and *Salmonella sp.* (■). Temperature (°C) (●). Hydrogen peroxide (▲). pH = 6.9.

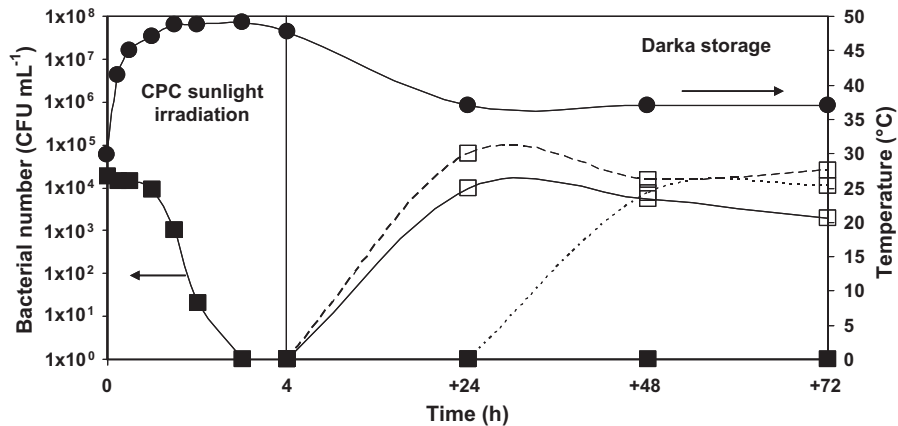


Fig. 6. Regrowth of *Salmonella sp.* after CPC solar treatment (experiment of Fig. 4a with differential subsequent storages). Well water of urban dam no. 1. (■) *Salmonella sp.* with sterile subsequent storage, (□) *Salmonella sp.* with non-sterile subsequent storage and 2 mg L⁻¹ of H₂O₂, (□-) *Salmonella sp.* with non-sterile subsequent storage and 5 mg L⁻¹ of H₂O₂, (□·) *Salmonella sp.* with non-sterile subsequent storage and 10 mg L⁻¹ of H₂O₂, temperature (°C) (●). pH = 6.9.

the addition of 10 mg L⁻¹ of H₂O₂ halted bacterial regrowth until 48 h. Low H₂O₂ concentrations (2 and 5 mg L⁻¹ of H₂O₂), when added just before storage, was no longer detected after 24 h, whereas 10 mg L⁻¹ of H₂O₂ were still present in low quantities (2 mg L⁻¹) and disappeared only after 48 h.

During storage, *Salmonella sp.* remains inactivated probably because of the addition of H₂O₂. Indeed, in presence of iron organo-complexes at natural pH, H₂O₂ could initiate a Fenton-like reaction producing ROS (Arslan et al., 2000; Lee et al., 2004). Nevertheless, the dark Fenton reaction is much slower and the NOM fraction competes with microorganisms for ROS consumption as observed in previous studies (Moncayo-Iasso et al., 2009; Rincón et al., 2001). When hydrogen peroxide is added, degradation of NOM occurs rendering it more bio-available and maintaining the bacterial charge. However, when H₂O₂ is exhausted, bacterial reactivation can occur when the

microorganisms are exposed to more favourable environmental conditions, via dark repairing processes as observed after illumination (Leyer and Johnson, 1993; Rincón and Pulgarin, 2004, 2007a). Indeed, bacterial persistence in *viable but non cultivable* phase (VBNC) and successive recovery is favoured because NOM is transformed into easily assimilated products. In this case, the addition of H₂O₂ after disinfection by CPC solar exposition should not be considered as a viable alternative to maintain water quality during successive storage, if NOM removal is not realized before.

4. Conclusion

For the first time, solar disinfection of large volumes natural water (18 L), comprising in 4 h of solar irradiation and the increase of temperature with natural surface water containing dissolved iron, showed a complete bacterial

inactivation of wild strains (total coliforms and *Salmonella* sp.) without regrowth during 72 h of dark sterile storage. Solar disinfection was still faster in highly NOM-charged water, showing the potential effect of photosensitizers and temperature in the process. Under lower increases of temperature (<45 °C), the addition of hydrogen peroxide (30 mg L⁻¹) at the beginning of the solar exposition with the same water source allowed the inactivation of both microorganisms, but regrowth was observed during dark storage. The fast exhaustion of hydrogen peroxide (less than 2 h) can explain bacterial recovery by increasing NOM bioavailability. Therefore, with a CPC solar photo-reactor, the induction of the photo-Fenton process ($h\nu/\text{Fe}^{2+}/\text{H}_2\text{O}_2$) during solar irradiation was ineffective in highly NOM-charged water, which causes competitive and fast oxidant consumption. After simple solar CPC disinfection, storage tests in uncleaned containers (in order to simulate real conditions) with different concentrations of hydrogen peroxide were not able to avoid bacterial regrowth, also because there was a fast exhaustion of the oxygen radicals. Hence, the induction of the Fenton ($\text{Fe}^{2+}/\text{H}_2\text{O}_2$) process during post-irradiation events in non-sterile conditions was not efficient in maintaining water quality.

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