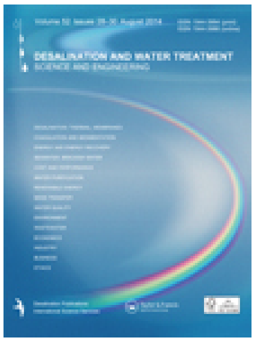

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Cost-effective and sustainable solutions to enhance the solar disinfection efficiency improving the microbiological quality of rooftop-harvested rainwater

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ABSTRACT

This paper investigates the efficiency of solar-based disinfection methods for improving the microbial quality of rooftop-harvested rainwater. Bacteriological water quality indicators including total and fecal coliforms, *Escherichia coli*, and Heterotrophic Plate Count were examined under different sunlight radiations. The efficiency of simple solar disinfection (SODIS) was enhanced at low pH values of the exposed rainwater. To enhance the concentrating effects of radiations, solar collector disinfection (SOCODIS) system was used. In addition, very simple and cost-effective techniques were employed to enhance the disinfection efficiency. This includes the wrapping of polyethylene terephthalate bottles with heat-resistant plastic bags to enhance the thermal/synergistic effects of radiations and the addition of natural acids (lemon/vinegar) for getting a low pH value of the rainwater in a natural way. Both simple SODIS and SOCODIS systems remained ineffective even under strong radiations and the best solution was to use the SOCODIS system with a combination of wrapping and addition of natural acids. A complete inactivation was achieved even at neutral pH by using reasonable concentrations of natural acids i.e. lemon/vinegar (0.5/0.4 ml) to avoid any taste/odor problem. Under moderate radiations, the same system was deemed best but at pH of 5. The only solution under weak radiations was to wrap the polyethylene terephthalate bottles by adding lemon/vinegar for obtaining a pH of 3 in the SOCODIS system.

Keywords: Drinking water; Lemon; Natural acids; Plastic bag; Point of use water treatment; Vinegar; Water disinfection

1. Introduction

There is a significant pressure on freshwater resources due to the increasing world's demand for the production of food, energy, utility goods, and services [1,2]. The access to a reliable and safe source of potable water is one of the major problems in developing countries and approximately one sixth of the world's population is facing this issue [3] and

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about 5,000 children die every day due to the water related problem of diarrhea [4]. The researchers suggest that improving the water quality at the household level, however, may reduce diarrhea by up to 40% [5–7]. So, there is a need to use the point-of-use water treatment methods at the household levels which are cost-effective and easy to use [8]. Rooftop-harvested rainwater can be regarded as an alternative source of potable/nonpotable water supplies [9–12]. The microbial quality of this supply, however, is of concern which may restrict the use of rainwater for potable purposes. Table 1 summarizes the review on the presence of both indicator organisms and potential human pathogens in rainwater harvested from rooftop in different parts of the world [13–24]. The presence of indicator organisms in stored rainwater or roof run-off reveals that harvested rainwater has to be treated especially when it is used for potable purposes.

Solar disinfection (SODIS) has shown to be an effective point-of-use treatment method for household water supplies [25–28]. In simple SODIS, water in polyethylene terephthalate (PET) bottles or other simple containers is exposed to sunlight for about 6–8 h and pathogens are inactivated by the synergistic effect of both temperature and sunlight radiations [29–31]. Simple SODIS practice has limitations including the insufficient disinfection at all weathers in addition to the small amount of treated water. To overcome the lacking of the scientific data, a detailed study on the effectiveness of solar based disinfection methods for the treatment of rooftop-harvested rainwater was performed [32–34]. The experiments were performed at different radiation conditions depending upon the sunlight irradiance. Rooftop harvested rainwater in PET bottles with different initial pH/turbidities and different backing surfaces/directions was exposed to direct sunlight at rooftop for about 8–9 h in simple SODIS [32]. The incomplete microbial inactivation even under strong radiations, however, required more exposure time and/or means to enhance the thermal/optical effects of the sunlight.

Solar collector disinfection (SOCODIS) system was used to enhance mainly the optimal effects of radiations. It was a simple wooden box covered with aluminum foil having a rectangular base and side open wings for reflecting the sunlight radiations. In SOCODIS system, the disinfection efficiency increased by about 20–35% compared with simple SODIS but the disinfection was still incomplete even under moderate radiations [34]. Finally, few simple and low-cost techniques were used to enhance the disinfection efficiency of both simple SODIS and SOCODIS system.

Table 1
Range (except where mentioned) of indicator microorganisms in harvested rainwater in different countries

Location	Fecal bacteria	Concentration (per 100 ml of rainwater)	Source [Reference]
USA, rural area	Thermotolerant coliforms	10–20	Stored rainwater [13]
England, semi rural	*E. coli*	0–53	Stored rainwater [14]
	Fecal streptococci	0–79	
London, urban area	*E. coli*	0–16,000	Roof run-off [15]
	Enterococci	0–680	
Australia, rural towns	*E. coli*	0–370	Stored rainwater [16]
Australia, urban area	Thermotolerant coliforms	Maximum 800; Mean 119	Stored rainwater [17,18]
Australia, rural, urban industrial	Thermotolerant coliforms	Mean counts 55–125	Stored rainwater [19]
Germany, urban	*E. coli*	Median 26; Maximum 410,000	Stored rainwater [20]
	Fecal streptococci	Maximum 410,000	
New Zealand	*E. coli*	0–111	Stored rainwater [21]
	Thermotolerant coliforms	0–840	Stored rainwater [22]
	Enterococci	0–4,900	
Scotland, rural area	Fecal streptococci	Geometric mean range 482–46,580	Roof run-off [23]
	Thermotolerant coliforms	Geometric mean range 383–3,627	
Denmark, urban area	*E. coli*	4–990	Stored rainwater [24]

This included the addition of commonly available natural acids (N_A) to increase the process efficiency by decreasing pH to a minimum acceptable level and wrapping the PET bottles with heat-resistant plastic bags for increasing the water temperature to enhance the synergistic effects of ultraviolet (UV) and infrared (Vis + IR) radiations. A series of experiments were conducted and the summary of the results/findings is presented in this study.

2. Materials and methods

2.1. Rainwater samples

All rainwater samples were taken from the underground storage tanks of a rainwater facility located at Seoul National University in Seoul, South Korea. The rough schematic diagram of the rooftop rainwater harvesting system and a detailed description has been published by Han and Mun [35].

2.2. Disinfection experiments

Sunlight irradiance measured over a period of about one year was categorized as three weather of equal number of months as weak, moderate, and strong weathers. Weak weather represents sunlight radiations of 220–450 W/m^2 with an average value of about 300 W/m^2, moderate weather represents sunlight radiations of 450–700 W/m^2 with an average value of about 550 W/m^2, and strong weather is represented by sunlight radiations of 650–>1,000 W/m^2 with an average value of about 850 W/m^2 for about four months from May to August. For simple SODIS and SOCODIS system, microbial inactivation was monitored in all weathers but results of only strong weather were presented (Fig. 1). Time 0 h corresponds to 8/9 am, when the irradiation of rainwater samples began while it ended at 5/6 pm, corresponding to 9 h.

Stored rainwater was exposed to direct sunlight under different radiations in 2 l PET bottles and the removal of all bacteriological parameters were monitored by analyzing the samples at regular time intervals of 1 h. Nontreated controls were maintained in the same environmental conditions but shielded from the sunlight by covering the PET bottles with aluminum foil and kept under room conditions. Under strong radiations, the samples were withdrawn after direct exposure to strong radiations for 8–9 h and were maintained at 26˚C (room conditions) for further 3 days along with the controlled samples in dark ("Control *E. coli*" in Figs. 1–5 refers to controlled samples for *E. coli*).

In simple SODIS, one used commercially available 2-l PET bottle with reflective backing (rear surface covered with aluminum foil) containing a 1.7 l rainwater sample was exposed to direct sunlight at the rooftop [32]. In a SOCO-DIS system, a simple box made of five wooden pieces, four covered with aluminum foil as side wings and one as a base containing four PET bottles were exposed to direct sunlight with each PET bottle containing 1.7 l of stored rainwater [34].

Locally available heat-resistant plastic bags, normally used to wrap new shirts, were also used to enhance the thermal and hence the synergistic effects by increasing the water temperature of the water inside PET bottles. These simple techniques were employed in both SODIS and SOCODIS system and the set of different system type and experimental conditions are presented in Table 3. Both lemon and vinegar were used as commonly available natural acids in different concentrations/ratios to reduce the pH values to 7, 5, and 3 for achieving the good disinfection efficiencies based on the earlier findings [32]. For a pH adjustment of 7, the used lemon concentration was about 0.5 ml (0.05% by volume) per liter of rainwater, while 0.4 ml (0.04% by volume) of vinegar was used to reduce the pH value of parent rainwater sample from 9–10 to the neutral pH value of 7. To reduce the pH value to 5, 1.5 ml (0.15% by volume) or 1.3 ml (0.13% by volume) of lemon or vinegar were used, respectively, while a pH 3 was obtained by adding 6 ml (0.6% by volume) or 4 ml (0.4% by volume) of lemon or vinegar, respectively, in 1 l of rainwater [33].

2.3. Measurements

Sunlight radiations were monitored on-site with a SP-110 Pyranometer (Apogee Instruments Inc., Logan, USA) connected to a datalogger (DT80 Series 2) recording 1 min averages in Watt/m^2 (W/m^2). Turbidity was measured using a turbidimeter (Hach 2100, USA), pH and water temperature were measured using a pH meter (Hach Sension 1, USA) and temperature probe, respectively, while dissolved oxygen and electrical conductivity were measured using the DO meter (Sension 378—Hach comp. USA).

Basic physicochemical parameters including pH and turbidity were also analyzed together with bacteriological parameters including total and fecal coliforms (FC), Escherichia coli (*E. coli*) and Heterotrophic Plate Count (HPC). The range of the different physicochemical/bacteriological parameters in parent rainwater samples monitored over a period of one year is given in Table 2. The pH of the rainwater samples was neutral and initial turbidities were low (<5 NTU).

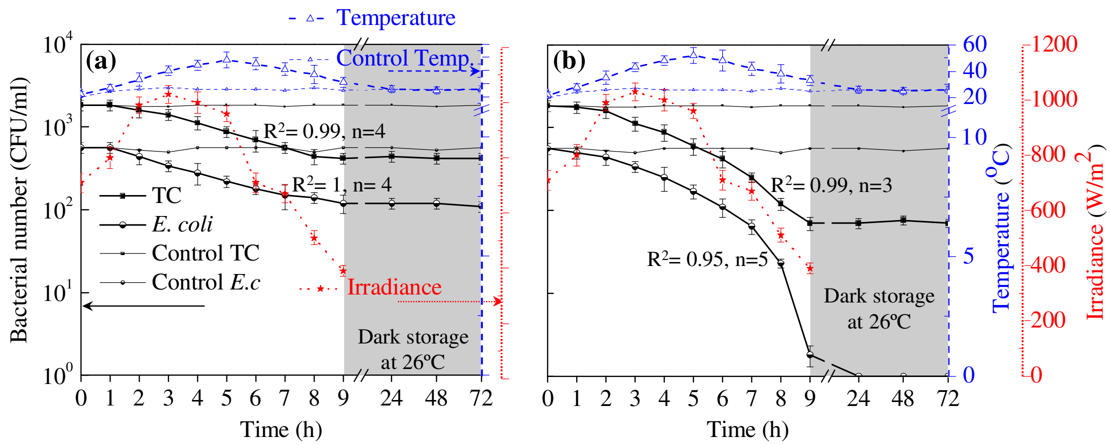

Fig. 1. Microbial inactivation under strong radiations in (a) SODIS and (b) SOCODIS system.

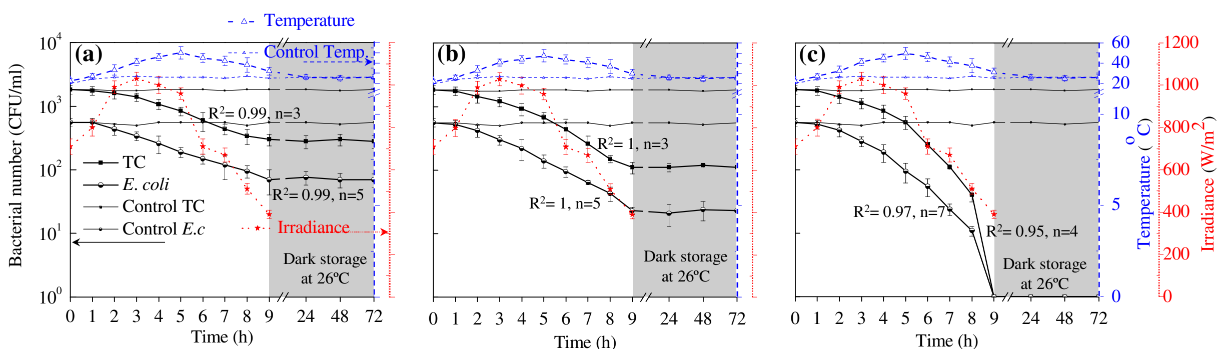

Fig. 2. Microbial inactivation under strong radiations in SODIS with (a) plastic wrapping, (b) natural acids, and (c) plastic wrapping & natural acids.

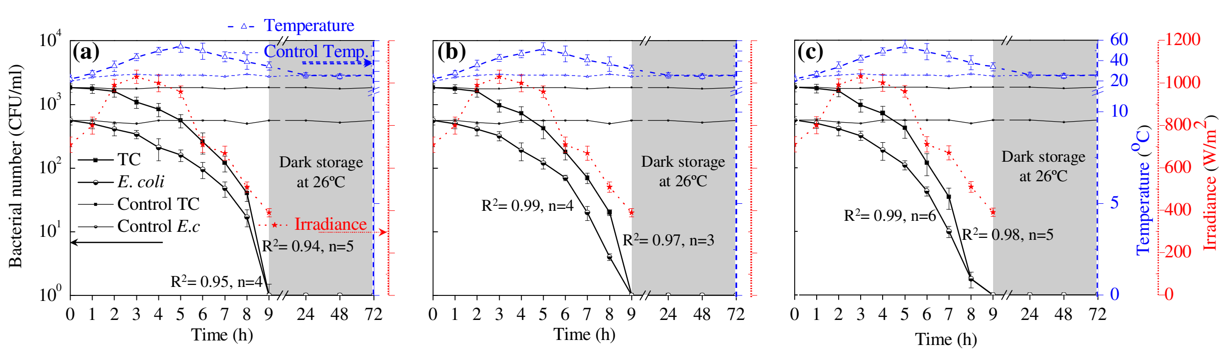

Fig. 3. Microbial inactivation under strong radiations in SOCODIS system with (a) plastic wrapping, (b) natural acids, and (c) plastic wrapping & natural acids.

Results are presented based on the mean average values at each point.

The water quality analysis was carried out in accordance with the guidelines described in the Standard Methods [36]. Both total and FC and *E. coli* were measured using the multiple tube fermentation technique. A series of fifteen test tubes with five tubes per each dilution of 10, 1, and 0.1 ml for each sample was used. Difco™ Lauryl Tryptose Broth (Becton, Dickinson and Company) was used for the presumptive phase at incubation temperature of 35°C for 24 ± 2 h or 48 ± 4 h. Positive tubes with growth (gas bubble or effervescence) were further subjected to confirmation phase at incubation temperature of 35°C for 24 ± 2 h or 48 ± 4 h.

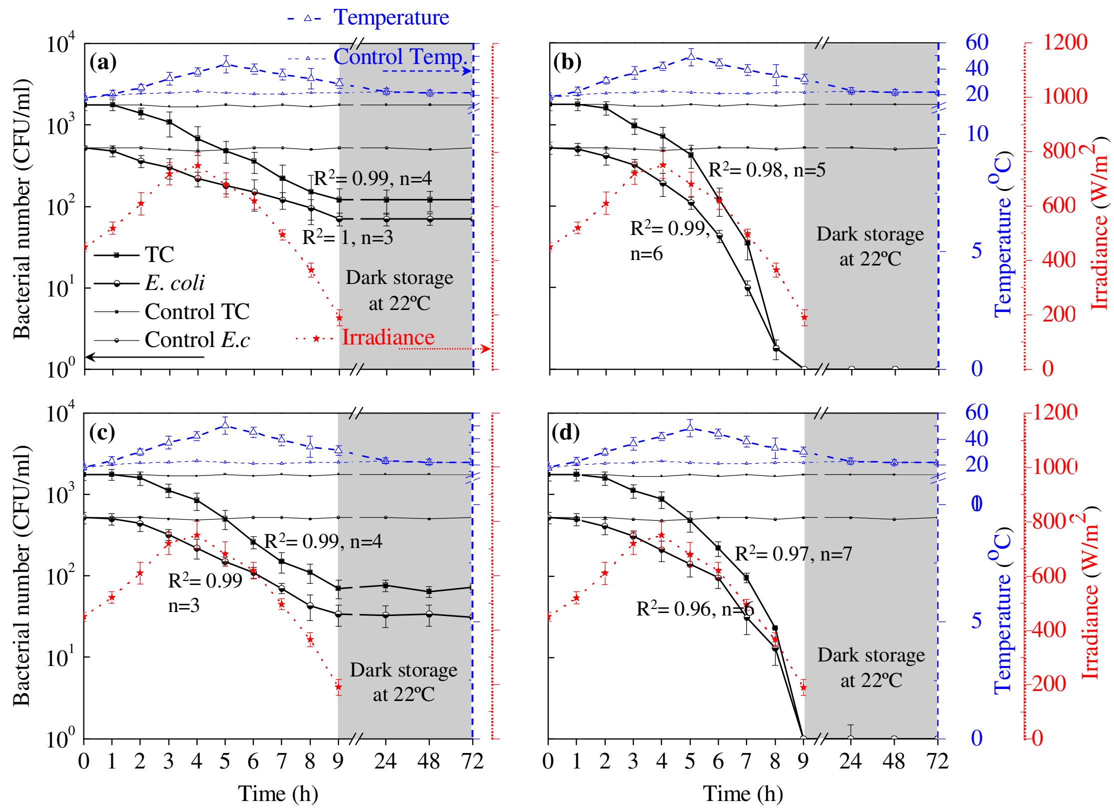

Fig. 4. Microbial inactivation under moderate radiations in simple SODIS with (a) plastic wrapping & natural acids and in SOCODIS system with (b) plastic wrapping & natural acids, (c) plastic wrapping, and (d) natural acids.

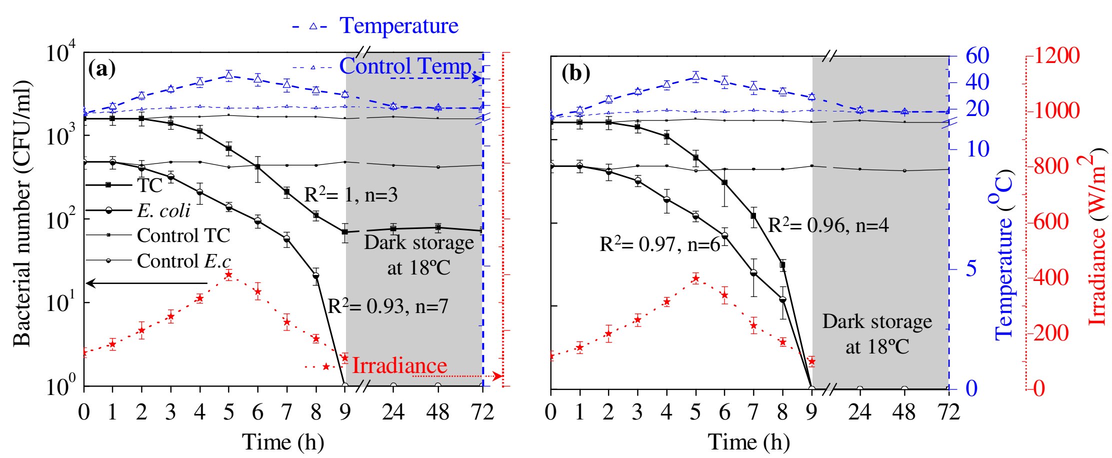

Fig. 5. Microbial inactivation under weak radiations in SOCODIS system with (a) natural acids, and (b) plastic wrapping & natural acids.

by using Difco™ Brilliant Green Bile Broth (Becton, Dickinson, and Company) and Difco™ EC Medium (Becton, Dickinson, and Company) for total coliforms (TC) and FC, respectively. Bacto™ EC Medium with Mug (Becton Dickinson France S.A.) was used for *E. coli* confirmation by incubating positive tubes at 44.5 ± 0.2˚C for 24 ± 2 h. For FC and *E. coli*, the incubation was done. Most probable numbers were recorded against the combinations of all positive tubes (gas production with growth, effervescence or yellow color, in case of TC and FC and bright blue fluorescence using a long-wavelength UV lamp in case of *E. coli*). The HPC was determined by the Pour Plate Method using Difco™ Plate Count Agar (Becton, Dickinson

Table 2
Physicochemical and bacteriological characteristics of parent rainwater samples

Parameter	Value
Temp. (°C)	18 ± 4
pH	8–10
Turbidity (NTU)	5 ± 3
Conductivity (μS/cm)	220 ± 70
Dissolved oxygen (mg/l)	10 ± 3
TC (CFU/100 ml)	1.6×103–1.84×10^3
E. coli (CFU/100 ml)	4.8×102–5.6×10^2
FC	6.5×10^2–1.1×10^3
HPC	2×103–2.7×10^3

and Company). A detailed description of the method is explained in authors' previous research [32].

2.4. Statistical analysis and modeling

Almost all experiments were repeated about 3–7 times to avoid any experimental error and the error bars in almost all the time graphs (Figs. 1–5) show the 90% confidence interval. The *R*-squared values are presented for validating the results based on the statistical criteria. Number of repetitions (*n*) and *R*-Square values for all types of experiments are shown in Figs. 1–5. Inactivation kinetics of all microbial parameters was investigated with the help of Gearaerd Inactivation Model Fitting Tool [37]. The inactivation rate constant (k_{max}, 1/min) along with standard error was evaluated and compared for both simple SODIS and SOCODIS system along with their modifications under different radiations (Table 4). The three models of Geeraerd (i.e. log-linear + tail, log-linear + shoulder, and log-linear + shoulder + tail) are widely used in SODIS scientific studies to fit experimental results [38]. The model was originally defined by coupling two differential equations as follows;

$$\frac{dN}{dt} = -k_{max} N \left(\frac{1}{1 + C_c}\right)\left(1 - \frac{N_{res}}{N}\right) \quad (1)$$

$$\frac{dC_c}{dt} = -k_{max} C_c \quad (2)$$

where "C_c" is related to the physiological state of the cells, "k_{max}" is the specific inactivation rate [1/time unit], and N_{res} is the residual population density [CFU/mL]. For the "log-linear + shoulder" model, which was the case in most of our experiments, inactivation model can be identified as,

$$N = N(0)e^{-k_{max}t} \frac{e^{k_{max}S_l}}{1 + (e^{k_{max}S_l} - 1)e^{-k_{max}t}} \quad (3)$$

For identification purposes the model can be reformulated as,

$$\log 10(N) = \log 10(N(0)) - \frac{k_{max}t}{\ln(10)} + \log 10\left(\frac{e^{k_{max}S_l}}{1 + (e^{k_{max}S_l} - 1)e^{-k_{max}t}}\right) \quad (4)$$

Table 3
Experimental types and conditions for low pH and enhanced UV/heating effects

Serial no.	System type	Parameters affecting disinfection	Description
1	SODIS	UV + IR	21 PET bottle with aluminum backing (PET_{Al})
2	W_P-SODIS	UV + IR + heat	PET_{Al} wrapped in heat-resistant plastic bag (W_P) for additional heating effects due to temperature increase
3	N_A-SODIS	UV + IR + low pH	PET_{Al} containing natural acids (N_A) i.e. lemon and/or vinegar for catalytic effects due to decreased pH
4	W_PN_A-SODIS	UV + IR + heat + low pH	W_P PET_{Al} containing N_A for additional heating and catalytic effects due to high temperature and low pH, respectively
5	SOCODIS	UV^* + IR	Several PET_{Al} in a rectangular wooden box with a base and side open wings covered with aluminum foil (SOCODIS) for enhanced radiation effects (UV^*) due to reflection
6	W_P-SOCODIS	UV^* + IR + heat	W_P several PET_{Al} in SOCODIS system for additional heating effects due to temperature increase
7	N_A-SOCODIS	UV^* + IR + low pH	Several PET_{Al} in SOCODIS system containing N_A for catalytic effects due to decreased pH
8	W_PN_A-SOCODIS	UV^* + IR + heat + low pH	W_P several PET_{Al} in SOCODIS system containing N_A for additional heating and catalytic effects due to high temperature and low pH, respectively

Table 4
The inactivation rate constant k_{max} (1/min) and the standard error (in parenthesis) for TC and *E. coli* analysis under different radiations and experimental conditions

Weather	System	Type	TC	*E. coli*
Strong	SODIS		0.26 (0.04)	0.55 (0.09)
		W_P	0.35 (0.04)	0.30 (0.03)
		N_A	0.51 (0.03)	0.50 (0.02)
		W_P N_A	1.85 (0.32)	1.10 (0.14)
	SOCODIS		0.62 (0.03)	1.40 (0.23)
		W_P	1.89 (0.33)	1.51 (0.25)
		N_A	1.57 (0.20)	1.29 (0.10)
		W_P N_A	1.66 (0.15)	1.26 (0.08)
Moderate	SODIS	W_P N_A	0.43 (0.03)	0.25 (0.02)
	SOCODIS	W_P	0.57 (0.04)	0.46 (0.03)
		N_A	1.72 (0.23)	1.38 (0.19)
		W_P N_A	1.66 (0.15)	1.27 (0.08)
Weak	SOCODIS	N_A	0.72 (0.04)	1.62 (0.32)
		W_P N_A	1.97 (0.27)	1.23 (0.15)

where "S_l" is a parameter represents the shoulder (time unit).

3. Results

3.1. Microbial inactivation in simple SODIS and SOCODIS system under strong radiations

The initial temperature of the parent rainwater sample was about 22˚C which increased to a maximum of 48 and 52˚C in SODIS and SOCODIS system, respectively, after 8–9 h of exposure to direct sunlight under strong radiations.

TC removal increased by 20% (percentage removal as compared with the initial microbial concentrations) in case of SOCODIS system when compared with that of SODIS (Fig. 1), as shown in Fig. 1. A 3-log inactivation (99.9%) in case of *E. coli* was observed in case of SOCODIS but TC inactivation was not complete (Fig. 1(b)). The inactivation difference between the weak and moderate weather and between the moderate and strong weather conditions (results are not shown due to similarity in inactivation trends) was about 20–25% and 30–40% in case of SODIS and SOCODIS system, respectively [32,34].

3.2. Microbial inactivation in modified SODIS and SOCODIS system under strong radiations

Both simple SODIS and SOCODIS system were modified in three different ways (Table 3) including the wrapping of PET bottles with heat-resistant plastic sheets (W_P-SODIS/SOCODIS), the addition of lemon/vinegar as natural acids in different ratio/ concentrations (N_A-SODIS/SOCODIS), and a combination of plastic sheets and natural acids (W_PN_A-SODIS/SOCODIS). The modifications were used even under strong radiations due to the incomplete inactivation at this weather. Figs. 2 and 3 represent the microbial inactivation of all the three cases in SODIS and SOCODIS system, respectively.

A 2–3˚C increase in water temperature inside PET bottles was observed and a maximum temperature of about 50 and 54.5˚C was recorded in W_P-SODIS and W_P-SOCODIS system, respectively (Figs. 2(a) and 3 (a)). A slight decrease of about 0.5–1˚C was observed in case of N_A-SODIS/SOCODIS system when compared with the simple SODIS/SOCODIS system (Figs. 2(b) and 3(b)). In case of SODIS, a 10% increase in TC/*E. coli* inactivation was seen after wrapping with plastic sheet (W_P-SODIS), most probably due to the enhanced synergistic effects of the thermal and UV radiations. None of the TC or *E. coli* was, however, completely inactivated even at the lowest effective pH value of 3 (initially adjusted by adding diluted HCl in parent rainwater sample), as shown in Fig. 2(a). A similar trend of increase in TC inactivation was observed in case of W_P-SOCODIS system and the modified system showed a 3-log inactivation (99.9%) for both TC and *E. coli* at adjusted pH value of 5 (Fig. 3(a)). The complete inactivation for all parameters including HPC was, however, possible only at the lowest initial adjusted pH value of 3.

The microbial inactivation was also improved after adding the natural acids i.e. lemon and/or vinegar in appropriate concentrations/ratios in the exposed rainwater to avoid any taste/smell. In N_A-SODIS, a pH value of 3 was adjusted by adding 6 or 4 ml of lemon or vinegar in 1 l of rainwater and the TC/*E. coli* inactivation is shown in Fig. 2(b). The microbial inactivation increased by about 17–18% for both TC and *E. coli* when compared with the simple SODIS system (Fig. 1(a)) but 2-log inactivation (i.e. 99%) was not achieved for any microbial parameter even at the lowest adjusted pH value of 3. For N_A-SOCODIS system, a complete microbial inactivation was achieved by adding the lemon/vinegar to adjust a pH value of 5 i.e. 1.5 or 1.3 ml of lemon or vinegar, respectively. A 3-log inactivation was achieved for both TC and *E. coli* after about 9 h (Fig. 3(b)) while both FC and HPC were also inactivated at this pH value and exposure time.

Finally, a combination of wrapping and addition of lemon/vinegar shows the complete inactivation in W_PN_A SODIS (Fig. 2(c)) when 6 or 4 ml of lemon or vinegar, respectively was used to adjust an initial pH value at 3. Temperature increase was about 2˚C due to wrapping, and a 3-log inactivation was obtained for

Table 5
Summary of the treatment results at all weathers in different systems

System type	Weather conditions			Reference
	Weak	Moderate	Strong	
SODIS			×	Fig. 1
SOCODIS			×	Fig. 1
W_P-SODIS			×	Fig. 2
N_A-SODIS			×	Fig. 2
W_PN_A-SODIS		×	✓	Figs. 2 and 4
W_P-SOCODIS		×	✓	Figs. 3 and 4
N_A-SOCODIS	×	✓	✓	Figs. 3, 4 and 5
W_PN_A-SOCODIS	✓	✓	✓	Figs. 3, 4 and 5

both TC and *E. coli* after 9 h of exposure (Fig. 2(c)). Both FC and HPC (4-log inactivation) were also completely inactivated. Similarly, in W_PN_A-SOCODIS system, the complete inactivation was obtained after 8–9 h of exposure time, as shown in Fig. 3(c). This was achieved by adjusting a neutral pH value i.e. by adding 0.5 or 0.4 ml of lemon or vinegar, respectively in 1 l of rainwater. Temperature increase was about 2˚C due to plastic wrapping which caused the temperature to remain above the critical value of 45˚C for more than 4 h for the thermal-optical synergistic effects. The complete inactivation of HPC was also achieved but after 9 h of exposure.

3.3. Microbial inactivation in modified SODIS and SOCODIS system under moderate radiations

Due to the incomplete inactivation under moderate radiations (450–700 W/m^2 with an average irradiance of 550 W/m^2) in both simple SODIS and SOCODIS system, all three modifications i.e. W_P, N_A, and W_PN_A were also employed at this weather for the SOCODIS system due to their effectiveness under strong radiations. In case of SODIS, however, the only effective modification under strong radiations i.e. W_PN_A was used under moderate radiations (Fig. 4(a)). The TC and *E. coli* inactivation is shown in Fig. 4 while the results for FC and HPC inactivation were not presented. The initial temperature of the parent rainwater sample was about 18˚C while the samples after 8–9 h of direct exposure to sunlight were maintained at 22˚C at room conditions for further 3 days along with the controlled samples in dark. Due to the plastic wrapping, the temperature difference was about 2–3˚C as compared with the simple SODIS and SOCODIS systems and the maximum temperature reached to about 44 and 49˚C in W_PN_A-SODIS and W_PN_A-SOCODIS system, respectively (Figs. 4(a) and 4(b)).

In W_PN_A-SODIS system, nearly 1-log inactivation was achieved i.e. about 93% for TC and 86% for *E. coli* (Fig. 4(a)) and it was 10–20% higher than the microbial inactivation under moderate radiations without any modification. The system, however, remained completely ineffective for the complete inactivation of any microbial parameter, as shown in Fig. 4(a) (results not presented for FC and HPC). In Fig. 4(b), the TC and *E. coli* inactivation is presented for the SOCODIS system for the W_PN_A case and a complete inactivation can be seen for both parameters after 9 h of exposure. The inactivation difference was about 15–25% compared with the SOCODIS system without plastic wrapping and addition of natural acids (TC and *E. coli* concentrations after 9 h of exposure reduced from 1,760 to 270 CFU/100 ml and from 520 to 68 CFU/100 ml, respectively, under moderate radiations in SOCODIS system). The synergistic effects might have played some role, unlike in W_PN_A-SODIS, due to a temperature increase to 45˚C and higher. The inactivation difference in both cases clearly indicates the effectiveness of adding the lemon/vinegar to lower down the pH value which was 3 and 5 in W_PN_A-SODIS and W_PN_A-SOCODIS system, respectively.

In W_P-SOCODIS system, nearly 95% inactivation was obtained for both TC and *E. coli*, as shown in Fig. 4(c) which was almost 15% higher than the simple SOCODIS system at this weather. Maximum temperature was about 50˚C with an increase of about 2˚C due to plastic wrapping. FC and HPC inactivation also increased by about 20% and 15%, respectively (results are not shown due to similarity in inactivation trends). The lowest initial pH of 3 (adjusted by adding diluted HCl) could be another reason for the enhanced inactivation other than the synergistic effects. In N_A-SOCODIS system, the same pH value of 3 was adjusted by adding lemon/vinegar, and a complete inactivation i.e. 3-log inactivation for both TC and *E. coli* was obtained after 9 h of exposure, as shown in Fig. 4(d). To adjust the pH values to 5 and 3, as mentioned earlier, 1.5 or 1.3 ml of lemon or vinegar and 6 or 4 ml of lemon or vinegar were added, respectively, in 1 l of rainwater.

3.4. Microbial inactivation in modified SODIS and SOCODIS system under weak radiations

The only possible modification for the effective disinfection at weak weather (220–450 W/m^2 with an average irradiance of 300 W/m^2) was to use the SOCODIS system with N_A and W_PN_A, since the system was not effective under moderate radiations with W_P. On the other hand, SODIS remained completely ineffective with any of the mentioned modifications under moderate radiations, so none of the modified SODIS system was tried under weak radiations. Microbial inactivation for the N_A-SOCODIS and W_PN_A-SOCODIS systems are shown in Figs. 5(a) and 5(b), respectively. The maximum temperature in W_PN_A-SOCODIS system was 44°C (Fig. 5(b)) which was slightly less than the critical value of 45°C required for the synergistic effects. In case of N_A-SOCODIS system, the maximum temperate was about 42°C, as shown in Fig. 5(a).

For N_A-SOCODIS system, a 3-log inactivation was achieved in case of *E. coli* but nearly 1.5-log inactivation was obtained for TC i.e. about 95% as compared with the initial microbial concentrations in the parent rainwater sample, as shown in Fig. 5(a). The inactivation difference, however, was about 25–30% as compared with the SOCODIS system without any addition of lemon/vinegar at this weather. FC inactivation also increased to about 95% but only 90% inactivation for HPC was achieved i.e. 1-log inactivation. 6 or 4 ml of lemon or vinegar, respectively were added to adjust initial pH value at 3. Same amount of lemon or vinegar was added in case of W_PN_A-SOCODIS system to adjust a pH of 3 and TC and *E. coli* inactivation is shown in Fig. 5(b). A 3-log inactivation was obtained after 9 h of exposure and a complete disinfection was achieved including FC and HPC. A 30% increase in the microbial inactivation was obtained in the modified W_PN_A system as compared with the SOCODIS system efficiency at this weather.

In Table 4, the inactivation rate constant i.e. k_{max} (1/min) and the standard error for all sets of experiments in both SODIS and SOCODIS system are presented for a quick analysis. Finally, Tables 5 and 6 summarize the findings of this study by highlighting both SODIS and SOCODIS system along with their modified forms (highlighted in Table 3) with plastic wrapping, addition of natural acids and a combination of both plastic wrapping and addition of natural acids.

4. Discussions

Microbial inactivation is directly related to sunlight intensity. All the results showed a similar tendency, signifying a close relationship between sunlight intensity and the time required to inactivate micro-organisms. An increase in rainwater temperature was due to the retaining of heat inside plastic bag due to which air temperature increases inside plastic bag which in turn heats up the rainwater inside PET bottles. The dependency of water temperature on radiations was observed and higher temperatures under strong radiations were recorded than under moderate radiations. Usually, a temperature difference of about 4–6°C was recorded between weak and moderate or between moderate and strong radiations. Also, the temperature varied during 8–9 h of exposures at all weathers and the highest temperature was recorded at about 1–2 pm when the radiations intensity on that particular day is at its peak.

Initial lag period showed persistent nature of micro-organisms against sunlight effects for about 1 h under strong radiations, while the lag was about 2–3 h under weak and moderate radiations (results not shown due to similarity of inactivation trends). Radiations effects were critical during middle stage when sunlight irradiance were high (>800 W/m^2) and the water temperature was also more than 45°C (Fig. 1 (b)). A direct correlation of radiation and inactivation was also observed when compared SODIS with the SOCODIS system under same radiations i.e. higher inactivation under weak, moderate or strong radiations in SOCODIS system then SODIS due to the enhanced radiations. Furthermore, the measured microbial concentrations until three days showed almost no re-growth or further inactivation due to residual effects of sunlight radiations, if any, in both SODIS and SOCODIS system. Also, the microbial recovery, as reported earlier, by means of some documented cellular repair mechanisms, such as photoreactivation [39] was not found.

Fig. 1 depicts that both simple SODIS and SOCODIS system remained ineffective for complete disinfection even at strong sunlight radiations, and microbial inactivation did not meet the potable guideline values i.e. 0 CFU/100 ml for TC, FC, and *E. coli* and 10 CFU/ml for HPC, however, the relative removal of indicator micro-organism was HPC < TC < FC/*E. coli*. It can be said that the microbial inactivation was mainly due to two mechanisms of treatment—thermal or pasteuri-

Table 6
Disinfection evaluation of different systems under different radiations

System type		1. SODIS			5. SOCODIS			Best solution	Alternate
		2. W_P-SODIS	3. N_A-SODIS	4. W_PN_A-SODIS	6. W_P-SOCODIS	7. N_A-SOCODIS	8. W_PN_A-SOCODIS		
Weather conditions	Strong	×	×	pH3	pH3	pH5	pH7	8	7 and 6 or 4
	Moderate	Nd	Nd	×	×	pH3	pH5	8	7
	Weak	Nd	Nd	Nd	Nd	×	pH3	8	NA
Remarks		Simple SODIS was not effective even at strong weather conditions			Simple SOCODIS system was not completely effective even at strong weather conditions			Best solution based on towards-neutral pH & simplicity of the system	

Note: × means incomplete disinfection, Nd: Not detected (evaluated), pHi: complete disinfection at pH value of "i".

zation—and UV radiations [40,41]. The synergistic effects of both can be seen due to fast inactivation of TC and *E. coli* in case of SOCODIS system, when the temperature was increased up to 45˚C and higher for about 4 h of exposure (Fig. 1(b)). Earlier findings also have suggested the synergistic effects when they are applied together [25] especially when the water temperature was 45˚C or above. This can be considered as a critical temperature beyond which either thermal or synergistic effects play an important role in disinfecting microbes.

By analyzing the effects of plastic wrapping on disinfection efficiency it can be said that temperature (thermal effects of sunlight) is not a predominant factor in microbial inactivation rather the UV + Vis. radiations determine the efficiency of the disinfection in addition to the lethal effects of low pH. It was also proposed that solar UV-A irradiation accounts for > 70% of the negative effects of sunlight [42]. UV radiations damage the nucleic acids of the cell or virus whose replication and transcription is inhibited by pyrimidine dimers and other photoproducts of nucleic acids and nucleic acid lesions [43]. It is also suggested that a likely cause of membrane dysfunction during UV-A irradiation in simulated sunlight is the damage to membrane enzymes [44]. The ability of cells to maintain the energy metabolism, in particular of the respiratory chain and their potential to generate adenosine triphosphate, is strongly affected even under a short exposure of about 30 min or less. The protein damage was also regarded as a crucial process during sunlight [45].

The acidification to pH 5 or below has a significant effect on disinfection rates, as was reported previously [46]. There are no health-based guidelines for pH; the 1996 Annual Report of the National Health and Medical Research Council indicates that the consumption of food or beverages with low (2.5) or high (11) pH does not result in adverse health effects [47]. As mentioned in the previous research [27], low pH values may have increased the inactivation rates due to the depletion of Adenosine triphosphate, the main energy storage and transfer molecule in the cells, and/or reducing equivalents due to the significant additional stress to the cells [48]. In addition, the lemon and lime juice concentrates possess intrinsic antimicrobial properties to eliminate *E. coli* and other bacterial pathogens at elevated temperatures [49].

It is recommended to replace/wash the used PET bottles in order to avoid the odor and taste problems due to the addition of lemon/vinegar. The practical benefit of using simple heat-resistant plastic bags and low-cost natural acids like lemon and vinegar in SOCODIS system will be the application of solar-based systems for the complete disinfection of stored rainwater even under weak radiations. Since, all the materials and techniques that are used to improve the efficiency of SODIS are simple, locally possible, and cost-effective, these innovative approaches can be easily applied at household and community scale in many parts of the developing world, especially in remote areas and where the centralized water supply system is not feasible, and thus contribute to achieve the Millennium Development Goals of the United Nations.

5. Conclusions

This study aimed at improving the efficiency of the solar-based disinfection methods by simple, low-cost and energy-efficient techniques. Three modifications were employed in simple SODIS and SOCODIS systems which include (1) the wrapping of PET bottles with heat-resistant plastic bags to enhance the thermal/synergistic effects by increasing the water temperature, (2) the addition of natural acids i.e. lemon and vinegar in appropriate concentrations/ratios for adjusting low pH values, and (3) a combination of wrapping and addition of lemon/vinegar.

At first, both simple SODIS and SOCODIS system were not effective even under strong radiations. SOCODIS system was, however, partially effective for completely inactivating both FC and *E. coli*. So, three modifications were employed under strong radiations for both simple SODIS and SOCODIS system. The best solution was to use the SOCODIS system with the combination of wrapping and natural acids, since the complete inactivation was possible even at neutral pH by using reasonable concentrations of lemon/vinegar (0.5/0.4 ml) to avoid any taste/odor problem. Alternatively, the N_A-SOCODIS system was the second choice but at pH of 5 with slightly higher concentrations of lemon/vinegar. Two more possibilities include the SOCODIS system with plastic wrapping or the SODIS with combined wrapping and natural acids under strong radiations. In both of these cases, however, pH of the product water was very low i.e. up to 3 with high concentrations of lemon/vinegar. Under moderate radiations, the best solution was the SOCODIS system in combination with wrapping and natural acids by adding either lemon or vinegar to adjust a pH of 5. The only alternative at this weather was the SOCODIS system by adding the natural acids in slightly higher concentrations to get an initial pH of 3. The only option under weak radiations was to wrap the PET bottles by adding lemon/vinegar for a pH of 3 in the SOCODIS system.

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