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EFFECTS OF UV BLOCKING AND HEAT-RESISTANT PLASTIC BAGS ON SOLAR DISINFECTION OF RAINWATER AT DIFFERENT WEATHERS

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Abstract

The efficiency of solar disinfection (SODIS) was evaluated and enhanced by concentrating sunlight at low pH values in a solar collector disinfection (SOCODIS) system at different sunlight intensities and is highlighted in the author's earlier findings. In order for further improving the rainwater disinfection, simple technique like wrapping the PET bottles with heat-resistant plastic bags (Wp) were used for enhancing the thermal/synergistic effects of solar radiations. An analysis of the PET bottle's samples showed increased absorbance with exposure time. The effect of ultraviolet (UV) radiations, as determined by blocking UV radiations, is a predominant factor in microbial inactivation and it is mainly the UV radiations which determine the efficiency of the disinfection. A microbial inactivation of about 20-30% in UV blocked Wp-SODIS/Wp-SOCODIS systems signifies the heating effects of solar radiations suggesting 45-50ºC as critical value above which thermal radiations (Vis+IR) contribute towards disinfection. The inactivation of total and fecal coliform, *Escherichia coli (E. coli)* and Heterotrophic Plate Counts (HPC) remained unchanged Wp-SODIS or Wp-SOCODIS system at weak weather. At moderate weather, however, HPC was the only inactivated microbial parameter in Wp-SOCODIS system. An increase of 5-6% in microbial inactivation in Wp-SODIS system at this weather suggests a temperature value of 40-45ºC beyond which synergistic effects play role in disinfection. Rainwater was disinfected completely at strong weather in a Wp-SOCODIS system while disinfection efficiency was enhanced by about 10% in Wp-SODIS due to a temperature increase of about 7°C. The analysis of the exposed and controlled rainwater samples over a period of 6 months showed no leaching of the harmful byproducts from PET bottles into stored rainwater samples.

Key words: disinfection, plastic bag, potable, radiations, rainwater

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1. Introduction

Rainwater harvesting (RWH), being an old water management practice (Handia et al., 2003; Qiang, 2003), has received increased attention worldwide as an alternative source of potable and non-potable water supplies (Amin and Han, 2009a; Baguma et al., 2010; Ghisi and Ferreira, 2007; Han, 2007; Hatibu et al., 2006; Heyworth et al., 2006; Ibrahim, 2009; Kim et al., 2005; Lee et al., 2010; Meera and Ahammed, 2006; Nazer et al., 2010; Spinks et al., 2006; Sturm et al., 2009; Zhang et al., 2010). However, its usefulness for potable purposes is limited because the quality of stored rainwater in domestic tanks is perceived as failing to meet drinking water quality standards. Sunlight might be

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able to provide a low-cost, sustainable, and simple method of treating contaminated drinking water in developing countries, where people have no access to alternative water treatment systems. Solar water disinfection (SODIS) is recommended by the World Health Organization (WHO) as a low-energy and cost-effective disinfection method (WHO/UNICEF, 2005). Among the household water treatment and storage (HWTS) methods, SODIS is used by less than 1% of the households throughout the developing world (WHO/UNICEF, 2011). However, effects of SODIS for rainwater disinfection aiming its use for potable puposes especially in developing countries have not been investigated.

The synergism of ultraviolet (UV) radiations and temperature plays an important role in SODIS for treating contaminated water (Ubomba-Jaswa et al., 2009). Batch scale solar-based treatment processes were used for the disinfection of small volumes of drinking water (Cooper and Goswami, 1998; Goswami, 1997). To achieve good water disinfection efficiency, simple SODIS was improved by the use of different backing surfaces in polyethylene terephthalate (PET) bottles, and different types of solar concentrators (Gelover et al., 2006; Kehoe et al., 2001). The absorptive materials were used, for example, blackened PET surfaces, to accelerate the rate of thermal inactivation of organisms by many researchers (Martin-Dominguez et al., 2005; Rijal and Fujioka, 2003; Sommer et al., 1997). These adsorptive surfaces, however, will be unable to raise the water temperature sufficiently at weak weathers.

For concentrating the solar radiations, aluminium foil was attached to the back of PET bottles, resulting an increased disinfection rate constant by a factor of 2 (Kehoe et al., 2001). For increasing the optical inactivation of sunlight, solar collector and reflectors were also used (Kehoe et al., 2001; Mani et al., 2006; Saitoh and El-Ghetany, 2002; Vidal and Diaz, 2000; Wegelin et al., 2001) and these contributions increased the amount of treated water in given exposure time. Rijal and Fujioka (2001) attributed the improved disinfection efficiencies due to increased water temperature in solar reflectors. Martin-Dominguez et al. (2005) used reflective solar boxes to reduce disinfection time to 3–4 h. The use of chemical additives such as citric acid and copper plus ascorbate with the addition of sodium percarbonate, lime juice/pulp, riboflavin etc have also been reported to enhance the efficiency of SODIS (Fisher et al., 2012; Harding and Schwab; 2012; Heaselgrave and Kilvington, 2010, 2011). *E. coli* reductions of □6 log units compared to 1.5 log units for standard SODIS in the same exposure time is reported when lime juice/pulp was used as additive (Harding and Schwab; 2012). Similarly, a 1.68 log10 inactivation enhancement, when compared with standard SODIS, was observed for cysts of Acanthamoeba castellanii in the presence of riboflavin (Heaselgrave and Kilvington, 2011).

The efficiency of sunlight was evaluated by using different sources of contaminated water, including wastewater, freshwater, seawater and bathing waters (Dan et al., 1997; Mascher et al., 2003; Sinton et al., 1999, 2002) and, recently, by using stored rainwater (Amin and Han, 2009b). Incomplete disinfection of rainwater, even under strong weather conditions and for an exposure time of about 8-9 hours, led to the idea of enhancing the thermal and optical effects of sunlight by the use of a simple wooden box with aluminum foil wrapped on open wings, termed solar collector disinfection (SOCODIS) (Amin and Han, 2009c).

In the SOCODIS system, the optical and thermal effects are enhanced because of the concentration of radiation after reflection by aluminum foil. The SOCODIS system exhibited 30 to 40% better dinfection efficiency than the SODIS and a complete disingection of rainwater was achieved both at moderate (under low pH) and strong weathers. At weak weather and to some extent at moderater weather (under neutral pH), however, the SOCODIS system was not effective in disinfecting stored rainwater.

In this study, the efficiency of this system was further enhanced by finding the ways to improve the thermal effects of SODIS and hence the synergistic effects of UV, visible and infrared (IR) radiations by obtaining the desired temperature. The objectives were to determine the contribution of different wavelengths on rainwater disinfection by using locally available UV-blocking sheets and to improve the disinfection efficiency by simple techniques like wrapping the PET bottles with locally available heat-resistant plastic bags aiming complete disinfection at all weathers.

2. Materials and methods

2.1. RWH system and rainwater characteristics

The water samples were taken from the underground storage tanks of a rainwater facility, installed in two buildings on campus at Seoul National University in Seoul, South Korea. The rough schematic diagram of the RWH system and a detailed description has already been published (Han and Mun, 2008). Basic physicochemical parameters, including pH and turbidity, of stored rainwater were analyzed together with bacteriological parameters, while the discussion was focused mainly on microbial inactivation during analysis. The water quality analysis was carried out in accordance with the guidelines described in the Standard Methods (APHA, 1999). References values of parent rainwater samples which were used in this study are shown in Table 1.

A natural pH value of about 7-9 with low initial turbidity of 2-6 NTU were recorded for the parent stored rainwater samples.

Table 1. Physicochemical and Bacteriological characteristics of parent rainwater samples

Physiochemical parameters	***Value***	***Bacteriological parameters***	***Value***
Initial Temp. *(^0C)*	20±3	TC *(CFU/100mL)*	1000±100
pH	8±1	FC *(CFU/100mL)*	375±60
EC *(µS/cm)*	200±80	*E. coli (CFU/100mL)*	250±30-
DO *(mg/l)*	6±2	HPC *(CFU/mL)*	1700±200
Turbidity *(NTU)*	4±2		

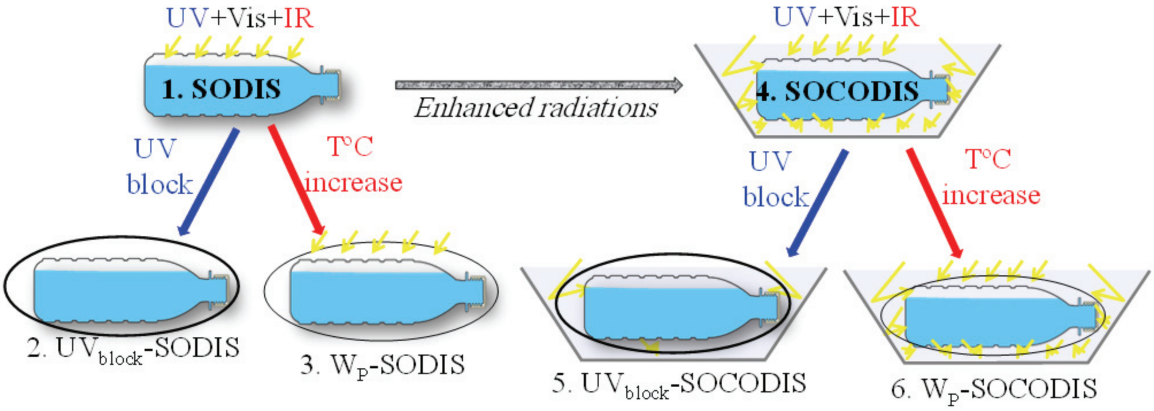


Fig. 1. Experimental description of both SODIS and SOCODIS system with plastic wrapping and UV-blocking setup
Note: W_P: plastic wrapping, UV_{block}: Blocking UV radiations

One main reason for different initial values of all physicochemical and microbial parameters, shown in Table 1, was the sampling time for all the experiments which is about one year and rainwater characteristics change due to different weather (winter and summer) and catchment conditions. Also, the storage conditions in underground concrete tanks changed the rainwater characteristics with time and with the addition of new rainfall in the tank. The different parent values have no effect; however, on experimental findings since same type of rainwater samples were used for one complete set of experiments in each case.

2.2. Description of solar-based systems

As shown in Fig. 1, a 2 liters PET bottle (Brand Name: MUHAK, Model number: WHITE WATER) containing about 1.7L of rainwater sample was exposed to direct sunlight for 8-9 hours with reflective backing in simple SODIS (Amin and Han, 2009b). Three PET bottles were places in each case for the triplicate analysis. In the SOCODIS system, four commercially available PET bottles were placed inside the box with open wings and exposed to direct sunlight for 8-9 hours at the rooftop of the building (Amin and Han, 2009c). The analysis was conducted throughout the year covering all the weather conditions. All PET bottles were kept undisturbed with an air space of about 15% of bottle volume during all experiments for air circulation to achieve the aeration. This was done since the solar-based water disinfection systems function effectively in fully aerobic conditions (Reed, 1997). Environmental and water temperatures were measured using a pH meter (Hach Sension 1, USA) placed beside the PET bottles at rooftop.

After the irradiation experiments in all weathers, rainwater samples were transferred into clean glass bottles, previously rinsed with bi-distilled water and stored in the dark at 4°C until analysis. Non-treated controls were maintained in dark (shielded from sunlight) by covering the PET bottle with aluminum foil kept under room conditions.

On-sight sunlight radiation was monitored on-site with a SP-110 Pyranometer (Apogee Instruments Inc., Logan, USA) connected to a datalogger (DT80 Series 2) recording 1 minute averages in Watt/m^2 (W/m^2). The UV MINDER® Model 3D-XP Xeroderma Erythemal SUV (Sunburning UV) intensity meter was used for measuring the UV-A and UVB spectrum. The meter can measure very low UV intensity up to 3 decimal places and measures UVB in Minimal Erythemal Doses per Hour (MED/hr) while UV-A light intensity was measured in milliwatts per square centimeter.

2.3. Microbial detection and modeling

Total coliform (TC), fecal coliform (FC) and *Escherichia coli (E. coli)* were measured using the multiple tube fermentation technique (MPN method) and the final concentrations were expressed as the number of colony-forming units (CFU) per 100 milliliter of original sample. Heterotrophic Plate Counts (HPC) was determined by the pour plate method and the final concentrations were expressed as the number of CFU per milliliter of original sample. Detailed descriptions of these methods have already been given (Amin and Han, 2009b). Bacteriological parameters were measured by taking approximately 40-45 mL of rainwater samples from each of the three and four PET bottles in SODIS and SOCODIS system, respectively. The samples were collected directly in polyethylene (50 mL) sterile screw cap containers, immediately chilled, and analyzed within 24 hours. The samples were taken at appropriate time intervals, usually after every 1.5 hours at both weak and moderate weathers and after each hour at strong weather, during 8-9 hours of exposure.

Most of the inactivation curves are based on the Geeraerd model, which explains the kinetics during mild-thermal inactivation processes, exhibiting a lag phase, followed by a log-linear phase, and then a tail of final concentration of remaining bacteria (i.e. shoulder + log-linear + tail). This model has been widely used in SODIS scientific contributions to fit experimental results (Geeraerd et al., 2000). In the present study, however, modeling with tailing was unlikely with the obtained data so most of the curves were modeled using only shoulder (a lag phase) and log-linear inactivation as is presented in Figs. 4, 6 and 7. This model has been also applied by the earlier researches on SODIS (Berney et al., 2006a, b; Bosshard et al., 2009; Sichel et al., 2007a, b)

Inactivation kinetics of all microbial parameters was investigated by using the Geeraerd Log linear and shoulder model in GInaFit and inactivation rate constant (k_{max}, 1/min) was evaluated and compared in both simple SODIS and SOCODIS system at all weathers and different experimental conditions i.e. when blocking UV and wrapping PET bottles with heat-resistant plastic bags. The Geeraerd model was originally defined by coupling two differential Eqs. (1-2).

$$\frac{dN}{dt} = -k_{\max}.N.(\frac{1}{1+C_c}).(1-\frac{N_{res}}{N}) \qquad (1)$$

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

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]. Following format of the model is used when actually applying to the experimental data in GInaFit.

$$log10(N) = log10\left[\begin{array}{l} (10^{log10(N(0))} - 10^{log10(Nres)}).e^{-k_{max}t} \times \\ \times\left(\dfrac{e^{k_{max}S_1}}{1+(e^{k_{max}S_1}-1).e^{-k_{max}t}} \right) + 10^{log10(N_{res})} \end{array} \right] \qquad (3)$$

Eqs. (1) and (2) can be obtained by substituting $S_{1,}$ a parameter represents the shoulder (time unit) or N_{res} equal to zero. The obtained values of k_{max} are shown in Fig. 4 for both SODIS and SOCODIS system when comparing the effects of UV blocking at strong weather. The k_{max} values in Figs. 6 and 7 represent only for the SODIS and SOCODIS system when wrapped with plastic bags and this is done for all four microbial parameters at moderate and strong weathers.

2.4. Effects of UV blockage and enhanced heating on disinfection efficiency

Locally available UV-blocking sheets were used to determine the effects of UV radiations on rainwater disinfection. For this purpose, a UV-absorbing plastic film was used for blocking the UV radiations reach the rainwater samples by wrapping PET bottles with this sheet. The transmitted light was measured and the amount of UV radiations (250-400nm) and visible light (400 to 700nm) was measured using a UV–vis spectrophotometer (Cary 50, Varian, CA). Both UV and visible light measurements showed that the used UV-blocking sheet absorbed almost 95% and 11% of the total UV radiations and visible light spectrum, respectively. The absorbance of the blocking sheets remained unchanged after one week of exposure to the sunlight. The UV transmittance, however, increased by 3-4% after one month exposure with a decreased transmittance of the visible light spectrum from 89% to 83%.

Simple heat-resistant plastic bags, normally used to wrap new shirts, were used to enhance the thermal and synergistic effects by increasing the temperature of the water inside PET bottles due to the trapped heat inside the plastic bag. About 0.15 mm thick plastic bags had about 92% and 88% transmittance of the solar radiations and infrared radiations, respectively. Each PET bottle was wrapped with this plastic bag in order to provide an air space of the same volume of the bottle i.e. approximately 2 liters. For determining the effects of UV radiations in sunlight and the enhanced heating due to heat-resistant plastic bags, six sets of experiments were performed in all weather conditions, as explained in Table 2, and the results are presented in the following sections.

The effects of UV radiations were investigated separately by using locally available UV-blocking sheets. Enhanced UV effects (UV*) are presented only in the SOCODIS system due to radiations reflection with open wings while enhanced heating effects (Heat*) are due to the use of heat-resistant plastic bags.

Non-treated 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. Only at strong weather, samples were withdrawn after about 9 hours exposure and the microbial concentrations were measured for further three days by keeping the exposed rainwater in dark along with control samples.

Table 2. Experimental types and conditions for UV and enhanced heating effects

S. no.	*Exp. type*	*Ray of light*	*Remark*
1	SODIS	UV + Heat	2 liters PET bottle with aluminum backing (PET$_{\mathrm{Al}}$)
2	UV$_{\mathrm{block}}$-SODIS	Heat*	PET$_{\mathrm{Al}}$ wrapped in UV-blocking sheet for blocking UV radiations reach the rainwater samples but wrapped in heat-resistant plastic bag for additional heating effects due to temperature increase
3	Wp-SODIS	UV + Heat*	PET$_{\mathrm{Al}}$ wrapped in heat-resistant plastic bag only for additional heating effects due to temperature increase
4	SOCODIS	UV* + Heat	Several PET$_{\mathrm{Al}}$ 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
5	UV$_{\mathrm{block}}$-SOCODIS	Heat *	Wrapping several PET$_{\mathrm{Al}}$ with UV-blocking sheet but also wrapped in heat-resistant plastic bag
6	Wp-SOCODIS	UV* + Heat*	Plastic wrapping of several PET$_{\mathrm{Al}}$ in SOCODIS system

At the end, nearly three to four samples of PET bottles, approximately 0.25-mm thick each, were cut out from the upper, middle and the lower smooth parts of the bottle from the side exposed to the sun. These samples were analyzed by UV-vis spectrophotometry to confirm the effects of the absorption properties of PET bottles on microbial inactivation.

Also, the UV-vis. absorbance was used to identify water quality changes in exposed and controlled rainwater samples. The sunlight irradiated (3 exposed PET bottles) and controlled (dark) rainwater samples were analyzed for 6 months (on monthly interval from July to December to cover all three weathers) for the concentrations of the plasticizers including di(2-ethylhexyl)adipate (DEHA) and di(2-ethylhexyl)phthalate (DEHP), formaldehydes and acetaldehydes. Solid-phase microextraction (SPME) technique was used and aldehydes were quantified by high-performance liquid chromatography (HPLC) after derivatization with 2,4-dinitrophenylhydrazine. The enriched fiber in the SPME method was placed in the injector port of gas chromatograph (GC, HRGC Mega 2 series, Fisons Instruments, Rodano, Italy). Finally, mass spectrometry (MS, Thermo Finnigan MAT, Bremen, Germany) was used for detecting the separated compounds.

3. Results and discussions

A solar spectrum has wavelengths in all ranges from less than 100nm to above 1400 nm but the radiation that reaches the earth and causes the disinfection includes mostly UV-A radiation, some parts of UVB radiation, and the visible range. The main fraction of solar ultraviolet radiation reaching the earth's surface i.e. UV-A light (320-400 nm) is believed to have both lethal and sublethal effects on *E. coli* (Berney et al., 2007). The heating effects due to solar radiations are also important, especially under moderate and strong weather conditions. Fig. 2 shows the absorbance characteristics of PET bottles used throughout the exposure experiments. The transmittance of used PET bottles was measured at strong weather in order to check the ageing of PET bottles after they had been used for one week (8-9 hours of exposure for 5 days), one month (8-9 hours of exposure for 20 days), and six months (8-9 hours of exposure for 120 days) by taking the unused PET bottle of the same kind as a reference.

The measured spectra of used PET bottles exhibited an increase in the absorbance of solar radiations in the UV-A range with increasing sunlight exposure time. The increased absorbance of the PET bottle after one month and six months, mainly at UV-A wavelengths, could be attributed to scratches on the surface partly caused by cleaning and reusing of same PET bottles for several experiments.

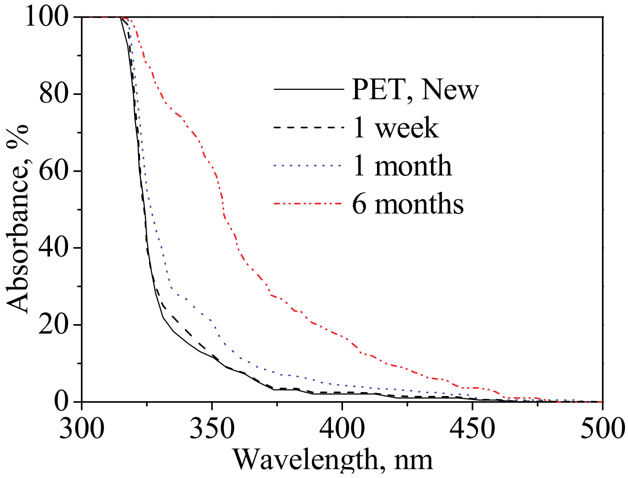


Fig. 2. Absorbance spectra of PET bottles with different exposure time

3.1. Radiation pattern at weak and moderate weather conditions

Weather was categorized into three different types, depending upon low, medium and high sunlight radiations specific to Seoul, South Korea (Latitude: 37°35' North, Longitude: 127° 03' East). Weak weather represents sunlight radiation of 100-400 W/m^2, with an average value of about 250 W/m^2 for approximately four months of the year, from November to February. Moderate weather represents sunlight radiation of 350-700 W/m^2, with an average value of about 500 W/m^2, during March, April, September, and October, and strong weather is represented by sunlight radiation of 650–>1000 W/m^2 with an average value of about 850 W/m^2 for about four months from May to August. These months of the year correspond to different weather conditions according to the weather pattern at experimental sites (Seoul, Republic of Korea). A generalized radiation pattern in weak and moderate weather conditions for one whole day's exposure time at Seoul is already published (Amin and Han, 2011) and is shown in Fig. 2. Each phase consists of increasing, nearly constant and decreasing radiations pattern with an equal time interval of about 3-4 hours, with the middle phase being critical for disinfection. The sunlight intensity of moderate weather is about double that of weak weather.

3.2. Temperature changes due to wrapping with plastic bags

Experiments were performed at all weathers and rainwater samples were taken, after every 1.5 hours at weak and moderate weathers while at every hour at strong weather, to measure the water temperature in simple SODIS and SOCODIS system with and without wrapping by heat-resistant plastic bags, as shown in Fig. 3 ('Wp' means wrapping with plastic bags). The data points represent the average of nearly six samples at strong weather only while in SOCODIS system; these are the average temperature values of all four bottles, which were kept inside the solar collector. A maximum difference of about 4-6°C can be observed between weak and moderate weather or between moderate and strong weather in SOIDIS or SOCODIS system. A temperature increase of about 3-4 and 4-5ºC was observed at weak and moderate weather, respectively, after wrapping with plastic bags in either SODIS or SOCODIS system. At strong weather, a maximum temperature increase of 7 and 9ºC was observed in SODIS and SOCODIS system, respectively after wrapping with plastic bags (Fig. 3c). The temperature increase due to plastic wrapping was the highest in strong weather but the difference was not significant between strong and moderate or moderate and weak weather. An increase in rainwater temperature was may be due to the retaining of heat inside plastic bag causing an increased air temperature inside plastic bag which in turn heats up the rainwater inside PET bottles.

The dependency of water temperature on weather conditions is obvious which increased in the beginning with increase in sunlight intensity and then started decreasing after about 5-6 hours with decrease in sunlight intensity usually in the afternoon.

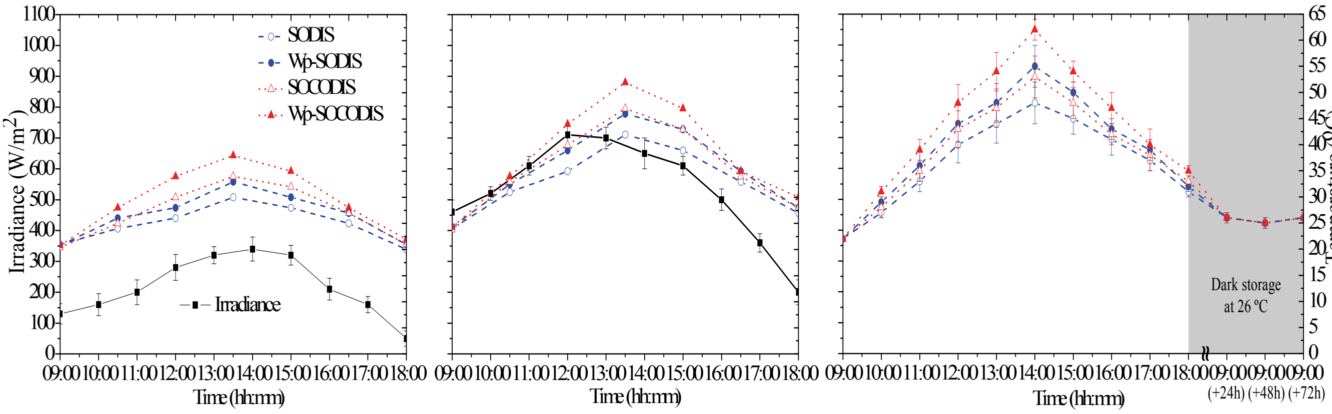
3.3. Contribution of UV radiation in water disinfection

In this section, an attempt is made to know the contribution of UV radiations and heating effects of sunlight towards disinfection. For this purpose, commercially available UV-blocking sheets were used along with the heat-resistant plastic bags in order to block the UV radiations and enhance the water temperature, respectively. Experiments were performed at all weathers but the results of only strong weather are presented in Fig. 4. The K_{max} (1/min) values in Fig. 4 represent the inactivation rate constants for both TC and *E. coli* with a corresponding coefficient of determination (R^2) of 0. 99 or 1, except where mentioned, calculated according to the Geeraerd model. A complete application of the model, both with tailing and shoulder, was not observed in most of the cases, and the absence of tailing could be considered due to the residual effects of sunlight radiation in later hours. In Figs. 4, 6 and 7, the data points represent the average of triplicate bacterial analysis and the error bars show the standard deviation of the mean.

As shown in Fig. 4, complete inactivation of both TC and *E. coli* was achieved in the SOCODIS system at strong weather (Fig. 4b) as compared with that of the moderate weather (results not shown). At this weather, samples were withdrawn after about 9 hours exposure and the microbial concentrations were measured for further three days by keeping the exposed rainwater in dark along with control samples, as shown by the shaded areas in Fig. 4.

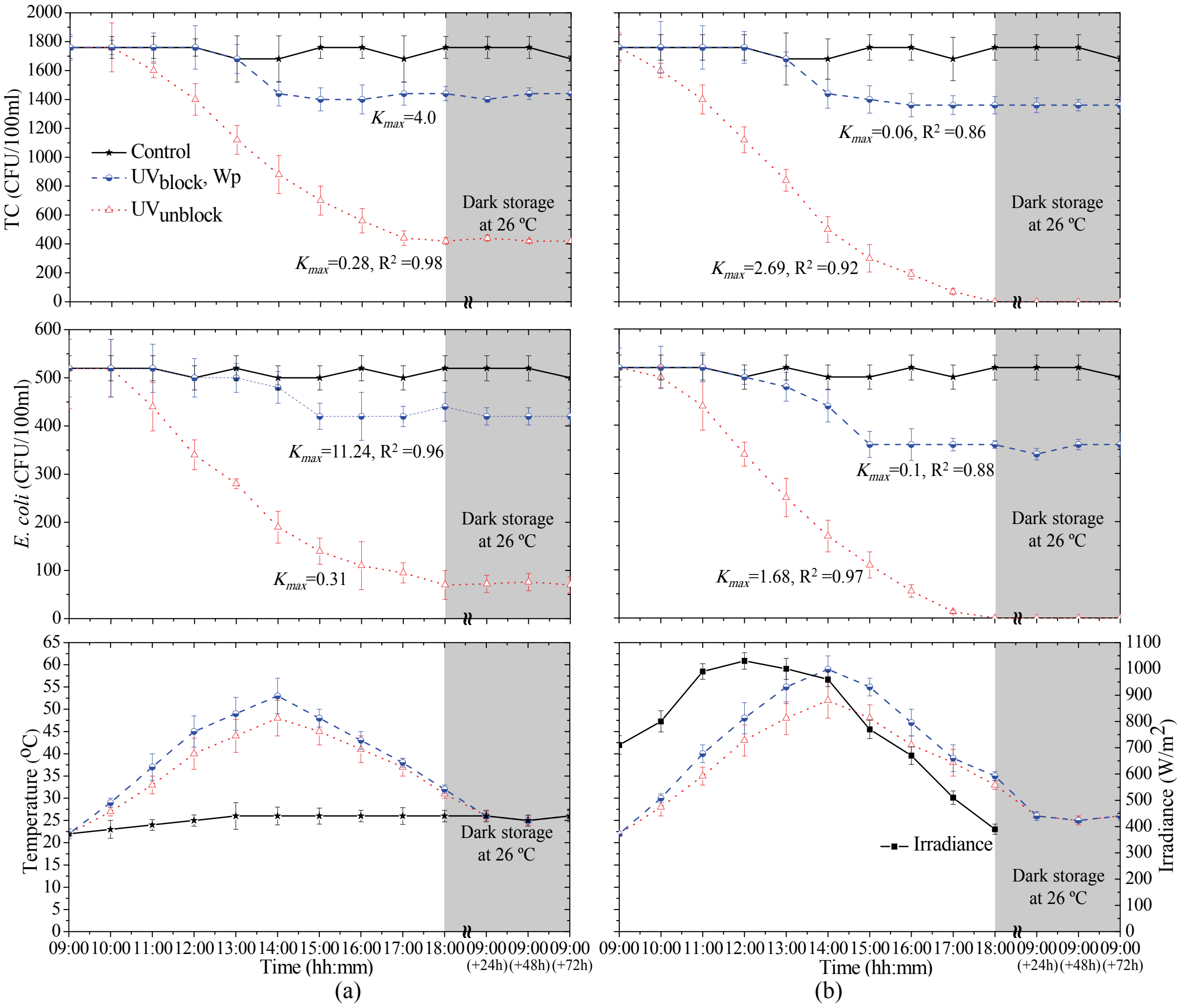
An insignificant yet interesting finding could be the temperature decrease by about 2°C after using the UV-blocking sheets and this could be understood by comparing the maximum temperature difference between Fig. 3c and 4b. A maximum temperature increase of 9ºC (Fig. 3c) was observed when only heat-resistant plastic bags were used while this value reduced to 7ºC after using the UV-blocking sheets with heat-resistant plastic bags (Fig. 4b). It is obvious from the results that SOCODIS system is more effective than SODIS and the inactivation difference was about 24% and 14% for TC and *E. coli*, respectively, when compared the observed microbial numbers with original concentrations. This inactivation difference reduced to about 5% and 12% for TC and *E. coli*, respectively, after using the UV-blocking sheets. Although, UV was blocked in both cases, however, an inactivation difference of about 5-12% between SODIS and SODODIS system signifies the effects of Vis+IR radiations i.e. heating effects due to higher rainwater temperature in SOCODIS system than SODIS (A maximum difference of about 7ºC as shown in Fig. 4 by comparing the dashed-blue lines for Wp-SODIS and Wp-SOCODIS system in Figs. 4a and b, respectively).

A microbial inactivation of about 20-30% in UV blocked Wp-SODIS/Wp-SOCODIS systems reveals that heating effects of solar radiations are present even at temperature below 50ºC. Another possible reason could be the amount of UV transmitted through these UV-blocking sheets which were capable of absorbing 95% of the total UV radiations. A temperature value of about 45ºC, however, can be considered as critical value above which thermal radiations (Vis+IR) contribute towards disinfection.



(a) (b) (c)

Fig. 3. Irradiance and temperature patterns at; (a) weak, (b) moderate, and (c) strong weathers



(a) (b)

Fig. 4. Effects of UV blocking on Temperature, TC and *E. coli* inactivation at strong weather in; (a) SODIS, (b) SOCODIS system

Microbial inactivation is directly related to sunlight intensity, which is obvious because of the strong optical effects of short wavelength components (i.e. 280–400 nm) of solar radiations, especially those in the UV-B range (280–320 nm) as is clear from an increase in the inactivation rate in the afternoon when UV-B is highest as a proportion of solar radiations (Fig. 5). The contribution of UV radiation at noon and in the afternoon was further evident from the UV strength radiation at these hours in our study, as shown in Fig. 5. In one of the studies, Wei et al. (1994) showed that longer wavelength radiation (>385 nm) is more effective in the inactivation of *E. coli*. Furthermore, unlike the general coliform group, *E. coli* is almost exclusively of fecal origin. Hence, the presence of non-fecal organisms in TC may be one of the reasons for the different behavior.

The higher UV strength in the afternoon (Fig. 5) could be one of the main reasons for microbial inactivation in the afternoon (Figs. 4, 6 and 7). The microbial inactivation after 5pm when UV strength almost reduced to zero (Fig. 5) could be attributed to thermal effects of sunlight radiations or the residual UV effects.

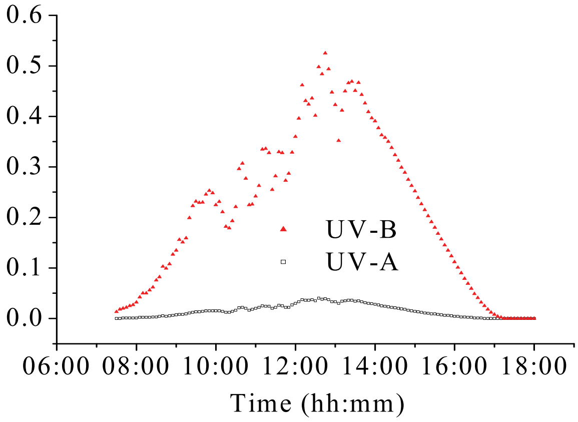


Fig. 5. UV strength pattern during a typical day at moderate weak weather at Seoul, South Korea. UV-B (280-320 nm) is expressed in MED/h and UV-A (320-400 nm) is expressed in mW/cm^2, while 1MED/h = 5.83x10^{-6}W/cm^2 (MED being the minimal erythema dose, i.e. the dose causing fair Caucasian skin to barely redden)

Results of Fig. 4, however, confirm that temperature (thermal effects of sunlight) is not a predominant factor in microbial inactivation and it is mainly the UV radiations which determine the efficiency of the disinfection. Acra et al. (1984) have proposed that solar UV-A irradiation accounts for >70% of the negative effects of sunlight.

Inactivation by UV is based on the damage caused to the nucleic acids (DNA/RNA) of the cell or virus. Primarily, the formation of pyrimidine dimers, but also that of other photoproducts of nucleic acids and nucleic acid lesions (von Sonntag et al., 2004) inhibits replication and transcription and thus prevents the cell or virus from multiplying. Bosshard et al. (2010a) have suggested that damage to membrane enzymes is a likely cause of membrane dysfunction during UV-A irradiation in simulated sunlight.

A short exposure of even less than an hour can strongly affect the ability of cells to maintain the energy metabolism, in particular of the respiratory chain and their potential to generate adenosine triphosphate. A reduction of carbon metabolism and defense against the oxidative stress is also noted during SODIS. Bosshard et al. (2010b) also have noted the protein damage as a crucial process in sunlight inactivation and have proposed an accelerated cellular senescence leading to the death of *E. coli*.

3.4. Effects of heat-resistant plastic bags on water disinfection

Simple heat-resistant plastic bags were used to enhance the thermal effect by increasing the temperature in both SODIS and the SOCODIS system. The analysis was performed in all weather conditions including the weak, moderate and strong weather aiming the complete disinfection, which was not achieved in the SOCODIS systems, especially for TC and HPC at moderate weather conditions (Amin and Han, 2009c). The contribution of the enhanced thermal effects on microbial inactivation due to the wrapping of PET bottles with heat-resistant plastic bags, for both SODIS and the SOCODIS system are shown in Figs. 6 and 7.

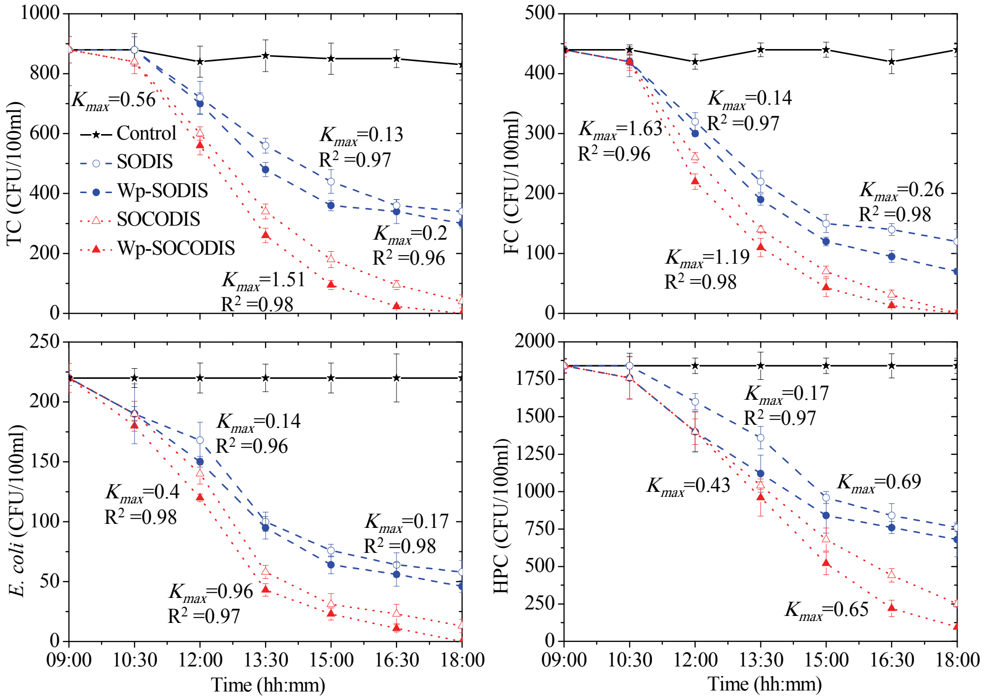
The maximum temperature difference between samples wrapped in plastic bags and an unwrapped sample was about 3°C and 4°C in SODIS and the SOCODIS system, respectively (Fig. 3) under weak weather conditions. An insignificant difference in the microbial inactivation between the unwrapped and wrapped samples (results not shown due to similar trends) could be due to the absence of the synergistic effects of the optical and thermal radiations since the maximum temperature was only about 33°C and 38°C in SODIS and SOCODIS system, respectively. These temperature values may not be enough to inactivate the microorganisms due to the synergistic effects let alone the thermal radiations since the rainwater temperature remained below the critical value of 50°C (Simon et al., 2007). It can be concluded, thus, that the thermal or even synergetic effects do not appear at temperatures below 40ºC. Figs. 6 and 7 represent the microbial inactivation of all parameters in both SODIS and SOCODIS system with and without plastic wrapping at moderate and strong weather, respectively. The K_{max} values in Fig. 7a are only for the Wp-SODIS and Wp-SOCODIS system since K_{max} values for SODIS and SOCODIS system for TC and *E. coli* at strong weather are already presented in Fig. 4.

At moderate weather, the temperature difference generated using plastic bags was about 5-6ºC in SODIS and SOCODIS system, thus not much different than the temperature increase at weak weather (Fig. 3). Disinfection efficiency improved due to temperature increase in both Wp-SODIS and Wp-SOCODIS system and the microbial inactivation increased by 5-6% for both TC and *E. coli* (Fig. 6b). Both TC and *E. coli* were completely disinfected in Wp-SOCODIS system. Usually, higher disinfection efficiency in SODODIS system was observed after wrapping with plastic bags, mainly because of a greater temperature increase in SOCODIS system compared with SODIS.

A complete disinfection was not achieved for any microbial parameter in Wp-SODIS while TC, FC and *E. coli* were disinfected completely in Wp-SOCODIS system. HPC was not disinfected completely (Fig. 7b) even in Wp-SOCODIS system although the final concentration was less than the drinking guideline of 100CFU/mL at moderate weather. In Wp-SODIS system, rainwater temperature remained above 40ºC for about 3-4 hours and above 45ºC for half of this time.

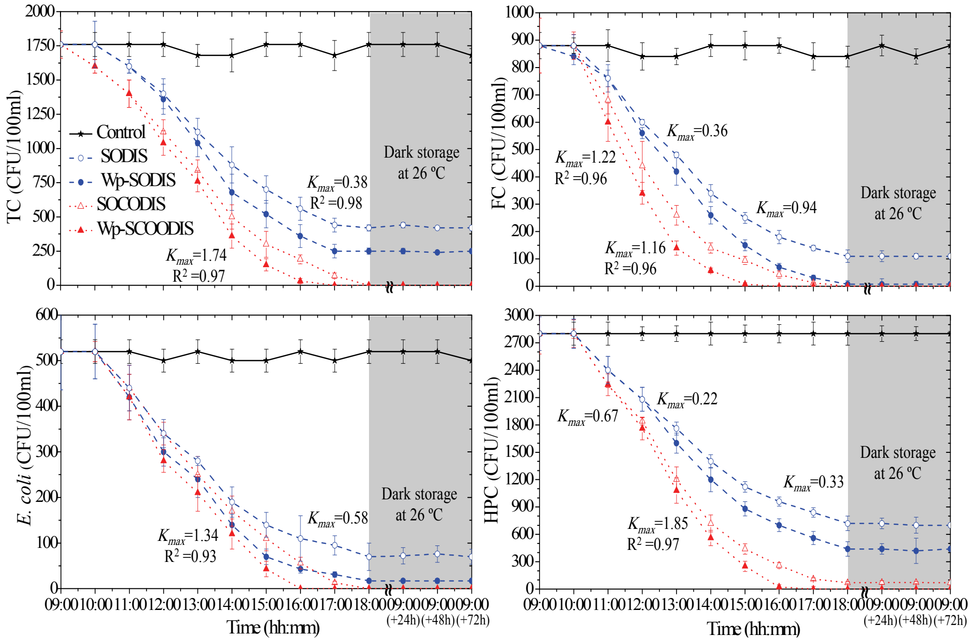
An increase of 5-6% in microbial inactivation in Wp-SODIS system reveals that the critical temperature beyond which synergistic effects play role in disinfection is somewhere between 40 and 45ºC. At strong weather, a complete microbial inactivation was achieved in case of SOCODIS system for all parameters including HPC when wrapped in heat-resistant plastic bags, as shown in Figs. 6c and 7c.

First of all, a maximum temperature difference of about 7 and 9ºC was observed in SODIS and SOCODIS system, respectively, after wrapping with plastic bags (Fig. 3c) due to an increased air temperature because of the trapped heat around the PET bottles and inside plastic bags. Nearly 10-12 % increase in the microbial inactivation in all microbial parameters was observed in SODIS after wrapping with heat-resistant plastic bag.



(a) (b)

Fig. 6. Effects of plastic bag's wrappings on microbial inactivation in SODIS and SOCODIS system at moderate weather



(a) (b)

Fig. 7. Effects of plastic bag's wrappings on microbial inactivation in SODIS and SOCODIS system at strong weather

This could be due to the increased thermal or synergistic effects of the solar radiations as the rainwater temperature remained above 45ºC for about 4-5 hours of the exposure time. The only inactivated microbial parameter, HPC, in case of SOCODIS system was also completely disinfected in Wp-SOCODIS system, most probably due to the detrimental effects of increased temperature. Other parameters like TC, FC and *E. coli* were also completely disinfected even after 7-8 hours of exposure time.

The initial temperature of the parent rainwater sample was about 22ºC at the strong weather. The samples were maintained at 26ºC in strong weather's conditions (room conditions) after exposure for further 3 days along with the controlled samples in dark, as shown by the shaded areas in Figs. 6c and 7c. The measured microbial concentrations until three days showed almost no re-growth or further inactivation due to residual effects of sunlight radiations in both SODIS and SOCODIS system.

Sunlight disinfection of rainwater, with or without plastic bags, exhibited three stages of treatment, depending upon the sunlight intensity, with the middle stage being critical. The initial lag period in the case of TC under weak weather conditions showed the persistent nature of non-fecal organisms against sunlight effects for a couple of hours or, more probably against UV attacks at low wavelengths.

HPC, however, showed prolonged resistance (results not shown) even in moderate weather conditions, in comparison with other parameters and the relative removal of indicator microorganism, observed in this study, was HPC < TC < FC / *E. coli*. It can be concluded from the present study that the microbial inactivation was due to two mechanisms of treatment i.e. UV radiations and thermal or pasteurization, which can work independently, but studies have indicated synergistic effects when they are applied together at temperature >45ºC (McGuigan et al., 1998). The thermal/synergistic effect started appearing during the afternoon, as can be seen in Figs. 6 and 7, when the temperature was 40ºC or above. So, a temperature of 40 and 45ºC can be considered as critical temperature values beyond which synergistic effects play an important role in disinfecting microbes.

3.5. Stability of PET bottles for long term use

The spectra of the exposed and controlled samples, as detected by UV-vis spectrophotometer were identical showing no release of UV-absorbing compounds from the PET surface into the rainwater samples. Table 3 shows the concentrations of DEHA, DEPA, and short-chain aliphatic aldehydes determined by triplicate analysis of the exposed rainwater samples. The concentration of individual aromatic aldehydes was always lower than the detection limit of 0.001mg/L.

Both acetaldehydes and formaldehyde content seems to correlate with exposure time. The maximum concentrations of both DEHA and DEHP after 6 months (120 days of exposure) were much lower than the WHO Guideline value for drinking water which is 80 and 8 μg/l for DEHA and DEPA, respectively. Identical compounds were detected in both exposed and controlled rainwater samples when analyzed with SPME-GC-MS.

So, it can be concluded that leaching of compounds from PET into rainwater samples is unlikely, at least, for six months of exposure. Finally, the results of this study have shown that with enhanced heating and radiations effects, the simple SODIS can prove effective for rainwater disinfection at all weathers. For moderate weather, only enhanced heating by using heat-resistant plastic bag may be effective for complete disinfection. These simple modifications with 8-9 hours of exposure time can also decrease the threshold value of global sunlight intensity (600–880 W/m^2 over a period of 6 hours) required for microbial inactivation (Berney et al., 2006c) which can be achieved in most developing countries (Martin-Dominguez et al., 2005). Among the other HWTS interventions, SODIS is the best solution for the people in developing countries with least financial resources. With $0.63 per person per year, SODIS remains the lowest cost household based intervention against waterborne disease.

The cost of flocculation/disinfection ($4.95), filtration ($3.03), or source-based interventions ($1.88 in Africa or $2.61 in Asia), or even chlorination ($0.66) are higher than SODIS (Clasen et al., 2007). SODIS, when compared with boiling, for example, can reduce costs for fuel used to boil water (McGuigan et al., 2012).

Table 3. Concentrations of DEHA, DEHP, Formaldehyde and Acetaldehyde in exposed rainwater samples for six months

Analysis time (month)	*No. of exposure days*	*DEHA (μg/l)*	*DEHP (μg/l)*	*Formaldehyde (mg/l)*	*Acetaldehyde (mg/l)*
0	0	0.005	0.008	0.001	0.001
1	20	0.008	0.09-0.15	0.003-0.006	0.005
2	35	0.01	0.16-0.2	0.01	0.007-0.009
3	55	0.016-0.020	0.18-0.25	0.024	0.01
4	80	0.021	0.3	0.02-0.028	0.008-0.012
6	120	0.037-0.04	0.8-0.9	0.041	0.01-0.015

The cost of the modified solar-based system by using the wooden box with wrapped aluminum foil (SODODIS system) and with the replacement of other inexpensive materials like heat-resistant plastic bags and PET bottles was estimated as 0.45-0.5$ per person per year.

4. Conclusions

This study aimed at assessing the feasibility of solar-based disinfection technologies of small quantities of potable water coupled with RWH to satisfy the daily needs of individuals or families in the rural/semi urban areas of developing countries.

The inefficiency of SODIS for rainwater disinfection in all weather and that of the SOCODIS system, at weak and moderate weather conditions, led to the idea of simple modifications to solar-based disinfection for complete disinfection. This includes the wrapping of PET bottles with simple, cost-effective and commonly available heat-resistant plastic bags that resulted in increased water temperature due to the retaining of heat and increased air temperature inside plastic bags.

The inactivation of investigated microbial parameters i.e. TC, FC, *E. coli* and HPC were monitored at all weathers in both SODIS and SOCODIS system along with the modified form when wrapped in plastic bags i.e. Wp-SODIS and Wp-SOCODIS system.

An analysis of the PET bottle's samples showed increased absorbance with exposure time mainly due to scratches on the bottle's surface requiring PET bottles to be replaced after using the bottles for a month or so. The use of UV-blocking sheets at strong weather revealed that the heating effects of solar radiations were present even at temperature below 50ºC. A microbial inactivation of about 20-30% in UV-blocked Wp-SODIS/Wp-SOCODIS systems may help to conclude 45ºC as critical temperature above which thermal radiations (Vis+IR) contributes towards disinfection.

The temperature increase after wrapping with plastic bags was not effective at weak weather both is SODIS and SOCODIS system as the critical temperature of 45°C, where thermal effects are considered effective for disinfection, was not achieved. An increase in the microbial inactivation of about 5-7%, however, could be attributed to the synergistic effects of thermal and optical radiations (i.e. UV and Vis+IR radiations) due to an increased temperature by 3-4ºC when wrapped with plastic bags. A complete disinfection was not achieved for any microbial parameter in either Wp-SODIS or Wp-SOCODIS system at this weather. The maximum temperature increase due to wrapping with plastic bags at moderate weather was about 5-6°C in SODIS and SOCODIS system, which enhanced the disinfection efficiency by about 5-6% in both Wp-SODIS and Wp-SOCODIS system. It may be concluded that the critical temperature beyond which synergistic effects play role in disinfection is somewhere between 40 and 45ºC. Rainwater was disinfected completely, with the exception of HPC, possibly due to the synergistic effects of thermal and optical radiations or due to the thermal effects alone as a result of increased temperature beyond 45°C for about 3 hours of exposure in Wp-SOCODIS system. A complete microbial inactivation including HPC was achieved at strong weather in Wp-SOCODIS system because of the thermal/synergistic effects of solar radiations due to an increased temperature of about 7-9ºC. At the same time, the inactivation time for FC and *E. coli* decreased from 9 to 7-8 hours. A 10% increase in the microbial inactivation in all microbial parameters was observed in Wp-SODIS due to the increased thermal or synergistic effects of the solar radiations as the rainwater temperature remained above 45ºC for about 3 hours of the exposure time.

Finally, the analysis of the exposed and controlled rainwater samples over a period of 6 months showed no leaching of the harmful byproducts from PET bottles into stored rainwater samples and hence these bottles can be used for long period of time like six months without any adverse health effects. The practical benefit of using simple heat-resistant plastic bags will be the application of solar-based systems for the complete disinfection of stored rainwater under moderate sunlight conditions to ensure potability in small-scale water-supply systems at the community level.

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