



Contents lists available at ScienceDirect

Desalination

journal homepage: www.elsevier.com/locate/desal

Improvement of solar based rainwater disinfection by using lemon and vinegar as catalysts

M.T. Amin^a, M.Y. Han^{b,*}^a Shaikh Mohammad Alamoudi Chair for Water Researches, King Saud University, Riyadh, Kingdom of Saudi Arabia^b Civil and Environmental Engineering Department, Seoul National University, Shinrimdong, Kwanak Gu, Seoul, 151–742, Republic of Korea

ARTICLE INFO

Article history:

Received 6 February 2011

Received in revised form 27 March 2011

Accepted 30 March 2011

Available online 17 April 2011

Keywords:

Lemon

Potable

Rainwater

Solar disinfection

Vinegar

ABSTRACT

The inefficiency of solar disinfection (SODIS) for the disinfection purposes can be improved by concentrating the sunlight i.e. by using solar collector disinfection (SOCO-DIS) system as investigated by authors in previous research or by increasing the exposure time of the targeted water to direct sunlight. Under weak sunlight conditions, however, this still remained ineffective as highlighted by earlier research. The efficiency of solar-based disinfection systems for the treatment of stored rainwater is further improved based on the better performance of these systems at low pH, by adding commonly available and inexpensive food preservative products, such as lemon and vinegar. Lemon and vinegar both increased the disinfection efficiency in SODIS by about 40% and completely disinfected rainwater in a SOCO-DIS system under weak weather conditions by lowering the pH to 3. An optimum combination of 2.5 ml (0.25%) of lemon and 1.7 ml (0.17%) of vinegar was selected to avoid any taste or odor problems while maintaining complete disinfection. Results showed that the choice of catalyst (lemon or vinegar in this study) was also an important factor in addition to low pH for disinfection using sunlight.

© 2011 Elsevier B.V. All rights reserved.

1. Introduction

Rainwater harvesting (RWH) can be a practical solution to water shortage [1,2], especially in developing countries, and is receiving increased attention worldwide as an alternative source of water supply as a result of climate change and increasing urbanization. Roof catchment is an established method of rainwater harvesting which has widely been used to provide urban populations with potable water supplies in many parts of the developing world [3–5]. RWH is considered crucial for meeting the future demand for potable water due to increased population and urbanization in developing countries, coupled with the recent evidence of climate change [6–9]. However, RWH has very limited use for potable water supply, the main reason being the quality of stored rainwater in domestic tanks, which is widely believed to be below drinking water quality standards.

Among the various methods of treating drinking water at the point of use to reduce exposure to microbial pathogens [10], solar disinfection (SODIS) is seen as a low-cost, sustainable, and simple method of disinfecting contaminated drinking water in developing countries where people have no access to alternative water treatment systems and need to use stored rainwater. Even those who have adequate supplies of water may not have access to microbiologically safe water, as improved supplies are often contaminated with pathogens that cause common

infectious diseases, such as cholera and enteric fever [11]. SODIS requires no commercial supply chain, as long as used polyethylene terephthalate (PET) bottles are available, and it has proved effective in significantly decreasing diarrheal diseases in children in developing countries [12,13]. PET bottles are considered safe and adequate for SODIS applications and may not cause any health risk [14] as no indication for migration of possible photoproducts or additives from PET bottles into water was observed during sunlight treatment of water [15].

Different approaches, such as different backing surfaces and solar concentrators, have been used to enhance the efficiency of SODIS [16–18] to achieve good water disinfection efficiency. Titanium dioxide (TiO₂) has been used as a catalyst both in suspended and immobilized forms to enhance the efficiency of SODIS disinfection in relation to fecal coliform (FC) [19] and *Escherichia coli* (*E. coli*) [20–22]. TiO₂ enhances the solar inactivation of *E. coli*, irrespective of whether it is used in particulate suspension or immobilized forms [23]. A simple batch-process solar photocatalytic disinfection system that uses a flat plastic sheet coated with TiO₂ is thought to be an appropriate and affordable technology for use in urban and semi-urban areas in developing countries [24]. An attempt is also made to accelerate the SODIS process by using a number of low-cost additives such as hydrogen peroxide, copper metal, and aqueous lemon and lime juice [25].

SODIS efficiency was previously evaluated using stored rainwater [14]. However, the attempted disinfection proved ineffective, even under strong weather conditions and for exposure times of around 8–9 h. The system was later modified by enhancing the thermal and optical effects of sunlight in a solar collector disinfection (SOCO-DIS) system

* Corresponding author.

E-mail addresses: mtamin@ksu.edu.sa (M.T. Amin), myhan@snu.ac.kr (M.Y. Han).

and disinfection increased by almost 30 to 40% compared with that of SODIS, thus enabling the disinfection of stored rainwater in strong and moderate weather conditions [26]. This improvement was mainly due to the concentration of radiation after reflection with aluminum foil and low pH adjustment. However, the SOCO-DIS system was not effective under weak weather conditions or, to some extent, under moderate weather conditions. While pH can be lowered in the laboratory using diluted HCl, this is not applicable in the field or for people at household level who require rainwater for potable purposes. Therefore, practical means are needed to lower the pH and hence achieve the same objectives as HCl in real situations.

To overcome the problems of incomplete disinfection under weak and moderate weather conditions, some simple techniques are used in this study, including addition of commonly available and cheap food products/preservatives to increase SOCO-DIS efficiency by decreasing pH to a minimum acceptable level.

2. Materials and methods

Non-treated controls were maintained in the same environmental conditions but shielded from sunlight by covering the PET bottle with aluminum foil kept under room conditions. Both lemon and vinegar were used as commonly available food products/preservatives to enhance the disinfection efficiency by decreasing the pH to around 3.

Basic physicochemical parameters including pH and turbidity were analyzed along with bacteriological parameters, though the discussion focuses mainly on microbial inactivation during analysis. The water quality analysis was carried out in accordance with the guidelines described in the Standard Methods [27]. Turbidity was measured using a Turbidimeter (Hach 2100, USA), pH and water temperature were measured using a pH meter (Hach Sension 1, USA), while DO and EC were measured using the DO meter (Sension 378 – Hach comp. USA).

2.1. Description of the RWH system

To perform the analysis, 2-L locally available, used PET bottles containing 1.7 liters of stored rainwater were kept on the rooftop of an engineering department building at Seoul National University campus in Seoul, Republic of Korea, as shown in Fig. 1. The rainwater in the bottles was taken from the underground storage tanks of an installed rainwater facility with detailed description as already published [14,26]. The bottles were shaken before exposure and left undisturbed during all experiments, with an air space of about 15% of bottle volume to allow for air circulation to achieve aeration [28].

2.2. Solar irradiation and description of SOCO-DIS system

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). In case of simple SODIS, one used commercially available 2-L PET bottle containing a 1.7-L rainwater sample and with reflective backing i.e. with an aluminum foil backing was exposed to direct sunlight at the rooftop [14]. 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 (Fig. 1) containing 4 PET bottles were exposed to direct sunlight with each PET bottle containing 1.7 L of stored rainwater [26].

2.3. Microbial detection and modeling

Total coliform (TC), FC and *E. coli* along with heterotrophic plate counts (HPC) were used as indicator organisms for the microbial quality of water. These were measured at appropriate time intervals, usually after every 2 h, during 8–9 h of exposure. TC, FC, *E. coli* were detected using the multiple tube fermentation technique (MPN method) and HPC was determined by the Pour Plate Method. A detailed description of the detection methods has already been published [14,26].

The inactivation curves are modeled and K_{\max} (1/min) was measured by using the Geeraerd Inactivation Model Fitting Tool (GInaFIT) [29] for testing three different types of microbial survival models on our data. The Geeraerd model explains the kinetics of mild-thermal inactivation processes exhibiting a lag phase, followed by a log-linear phase, and then a tail of the final concentration of remaining bacteria (i.e., shoulder + log-linear + tail). The three models used were: log-linear + tail, log-linear + shoulder, and log-linear + shoulder + tail [30] and these models have been widely used in SODIS scientific studies to fit experimental results [30]. All three models were run for each inactivation curve to compare the values of the Root Mean Sum of Squared Errors (RMSE) and finally the smallest RSME was selected as best fit for the respective inactivation curve.

2.4. Lemon and vinegar as food products

The PET bottles in the SOCO-DIS system, each with 1.7 L of stored rainwater and different concentrations of lemon and vinegar, were exposed to whole day sunlight under weak weather conditions. Three different pH values were adjusted using different concentrations of lemon and vinegar in each case and Table 1 gives a detailed description of the sampling conditions.

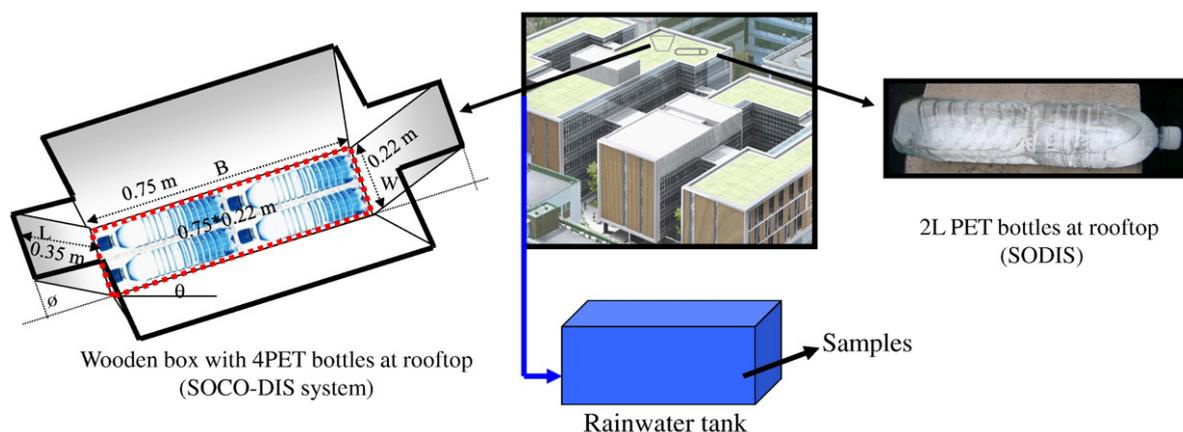


Fig. 1. Diagram showing the rainwater samples exposed to direct sunlight at rooftop in SODIS and SOCO-DIS system.

Table 1
Sampling conditions with different lemon and vinegar concentrations.

Sample type	pH	For 1 L rainwater		System
		Lemon/Vinegar (ml)	% Volume	
1	≈8	0/0	0/0	Parent rainwater
2	7	0.4–0.7/0.15–0.4	≤0.07/≤0.04	SODIS
3	5	1.5–2.1/0.7–1.3	≤0.2/≤0.13	
4	3	6.3–8/3.3–4	≤0.8/≤0.4	
5	7	0.4–0.8/0.2–0.4	≤0.08/≤0.04	SOCO-DIS
6	5	1.5–2.5/0.7–1.5	≤0.25/≤0.15	
7	3	7–8.5/3.5–4.2	≤0.85/≤0.4	

Table 2 presents a summary of the experimental conditions based on which the results are analyzed and discussed in the following sections. Most of the analysis was performed under weak weather conditions, except for the last case where a general comparison of SODIS and SOCO-DIS and comparison to gauge the respective efficiency of lemon and vinegar were performed under moderate weather conditions.

3. Results and discussions

3.1. Rainwater characteristics

Stored rainwater has a natural pH value of about 8, with low initial turbidity of <5 NTU and normal room temperature of about 24 °C. These high pH values in the stored rainwater could be due to the effects of newly constructed concrete catchment surfaces or due to the reinforced concrete storage tank. The microbial contamination of the stored rainwater is most probably due to the catchment surfaces which are comprised of both concrete and green roofing (ordinary grass) as well as a terrace catchment. These microbial concentrations decreased considerably once the catchment surfaces were cleaned (results are submitted by the authors for publication in other journal). The microbial quality improvement was about 98% for TC, FC and HPC while *E. coli* were removed completely after the first shower of rainfall. TC decreased to a final value of about 30–35 CFU/100 ml from an initial concentrations of 1500–2000 CFU/100 ml, FC decreased from about 900–1850 CFU/100 ml to about 20–25 CFU/100 ml while HPC decreased to about 150 CFU/ml from an initial value of about 6500 CFU/ml after the roof catchment was cleaned with the first shower of rainfall.

One of the reasons for the different initial values obtained for physicochemical and microbial parameters, as shown by error bars in Fig. 2, is the sampling time of the experimental analysis of around 6 months, during which time the weather conditions changed and maintenance of the catchment surfaces was carried out. Furthermore, the storage conditions in the underground concrete tanks changed the rainwater characteristics with time and with the addition of fresh rainwater into the tank. Microbial quality improved with time mainly due to the sedimentation effects inside the tank whereby the quality at the point of supply (situated at about 1.35 m from the base of the tank) was better than the microbial quality of the rainwater samples taken from the bottom of the tank. TC and FC concentrations were almost 45

Table 2
Sampling conditions with different lemon and vinegar concentrations.

Description	Testing parameter	Weather condition	pH adjusted by	System
Disinfection by adding vinegar	TC and <i>E. coli</i>	Weak	Vinegar	SODIS and SOCO-DIS
Disinfection by adding lemon	TC and <i>E. coli</i>	Weak	Lemon	SODIS and SOCO-DIS
Disinfection comparison between lemon and vinegar	HPC	Weak	Lemon and Vinegar	SOCO-DIS
Combined effects of lemon and vinegar	TC, <i>E. coli</i> and HPC	Weak	Lemon and Vinegar	SODIS and SOCO-DIS
Comparison of food products with simple SoDis and SoCoDis system	<i>E. coli</i>	Moderate	Lemon and Vinegar	SODIS and SOCO-DIS

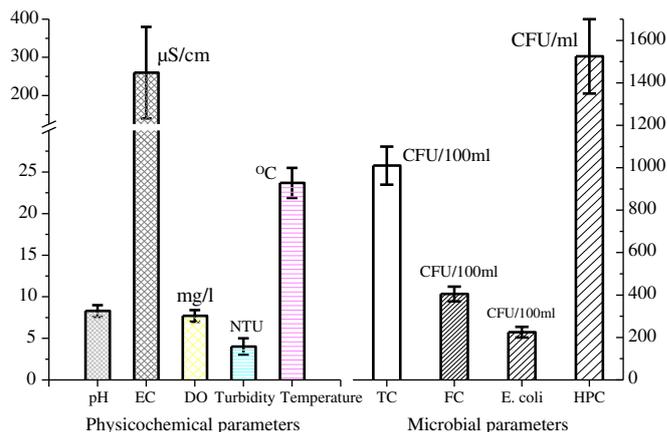


Fig. 2. Reference values for physicochemical and microbial parameters of parent rainwater samples.

and 30% less at the point of supply than at the bottom of the tank, respectively while this difference was about 75–82% for both *E. coli* and HPC (results not shown). A new rainfall event and hence the addition of newer rainwater into the tank increased the microbial concentrations inside tank due the contamination from the catchment surfaces. However, these initial differences did not affect the experimental analysis, as the same rainwater samples with different adjusted pH values were used for one complete set of experiments in each case.

3.2. Weather (irradiance and water temperature) characteristics

The solar collector was kept with an inclination to the mid-day position of sun. Irradiance and water temperature values corresponding to different weather conditions, specific to the weather patterns in Seoul, Korea (Latitude: 37°35' North, Longitude: 127° 03' East), are shown in Fig. 3.

Irradiation of the rainwater samples began at about 8–9 am, corresponding to 0 h in Fig. 3, while exposure to sunlight ended at 5–6 pm, corresponding to 9 h in Fig. 3. Weather conditions were categorized as weak, moderate or strong, depending on the sunlight radiations, which range from around 50 W/m² and less to more than 1000 W/m². In this study, weak weather conditions were defined as an irradiance range of 100–400 W/m² with an average value of around 250 W/m². Moderate weather conditions were defined as an irradiance range of 350 to 700 W/m² with an average value of around 500 W/m² of sunlight intensity. Strong weather is characterized by sunlight radiation of 650–1000 W/m² with an average value of about 850 W/m². A radiation difference of about 250 W/m², or almost double, is observed between weak and moderate weather conditions, as shown in Fig. 3.

The recorded temperatures in Fig. 3 are the representative results of around 5–7 repetitions, with error bars showing minimum and maximum values. For the SOCO-DIS system, the average temperatures were recorded for all the 4 PET bottles inside the wooden box. A maximum difference of 5 and 7 °C was observed between weak and moderate weather conditions in the case of the SODIS and the SOCO-DIS system, respectively. Temperature increase is dependent on sunlight

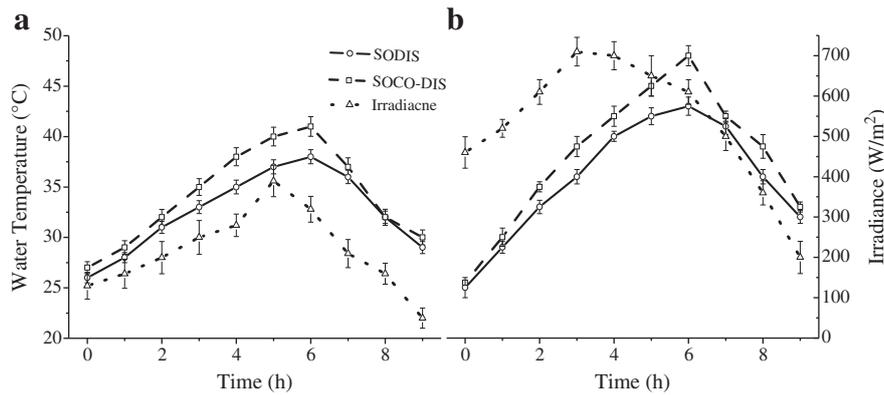


Fig. 3. Irradiance and temperature changes in the SODIS and SOCO-DIS systems with exposure time at (a) weak and (b) moderate weather conditions.

irradiance and increased with increasing sunlight radiation from morning to afternoon and then decreased until evening with the decreasing intensity of sunlight. Furthermore, the temperature difference between the SODIS and the SOCO-DIS was 3–5 °C due to the concentrated effects of sunlight radiation and reflection in the later case.

SODIS works on the basis of two major facts, the lethal action of solar UVA light, and the synergistic effect which is created when water temperature rises above 50 °C [31]. Unfortunately, it is not always possible to reach such high water temperatures inside the PET bottles under weak weather conditions.

3.3. Effects of low-cost food products

The effects of safe, readily available, and inexpensive food preservative, such as lemon and vinegar, with a target of decreasing the pH

value, were evaluated in relation to disinfection efficiency under weak weather conditions. The objective was to achieve complete disinfection at low pH using vinegar in a quantity that does not cause smell or taste problems. The main reason for using weak weather was the inefficiency of the SOCO-DIS system in this weather for all of the microbial parameters and, to some extent, for TC and HPC under moderate weather conditions [26]. In most of the cases, the results for FC and HPC were not presented because of similar inactivation trends to those of TC and *E. coli*.

The parent rainwater sample has a pH value of usually around 8 and above, but not more than 9. The corresponding vinegar concentrations for the same pH adjustment were almost half that of the lemon concentrations, as is clear from Table 1. There is no health-based guideline for pH in drinking water quality guideline (WHO, USEPA, etc.) although the pH range that is recommended is to avoid the corrosion in

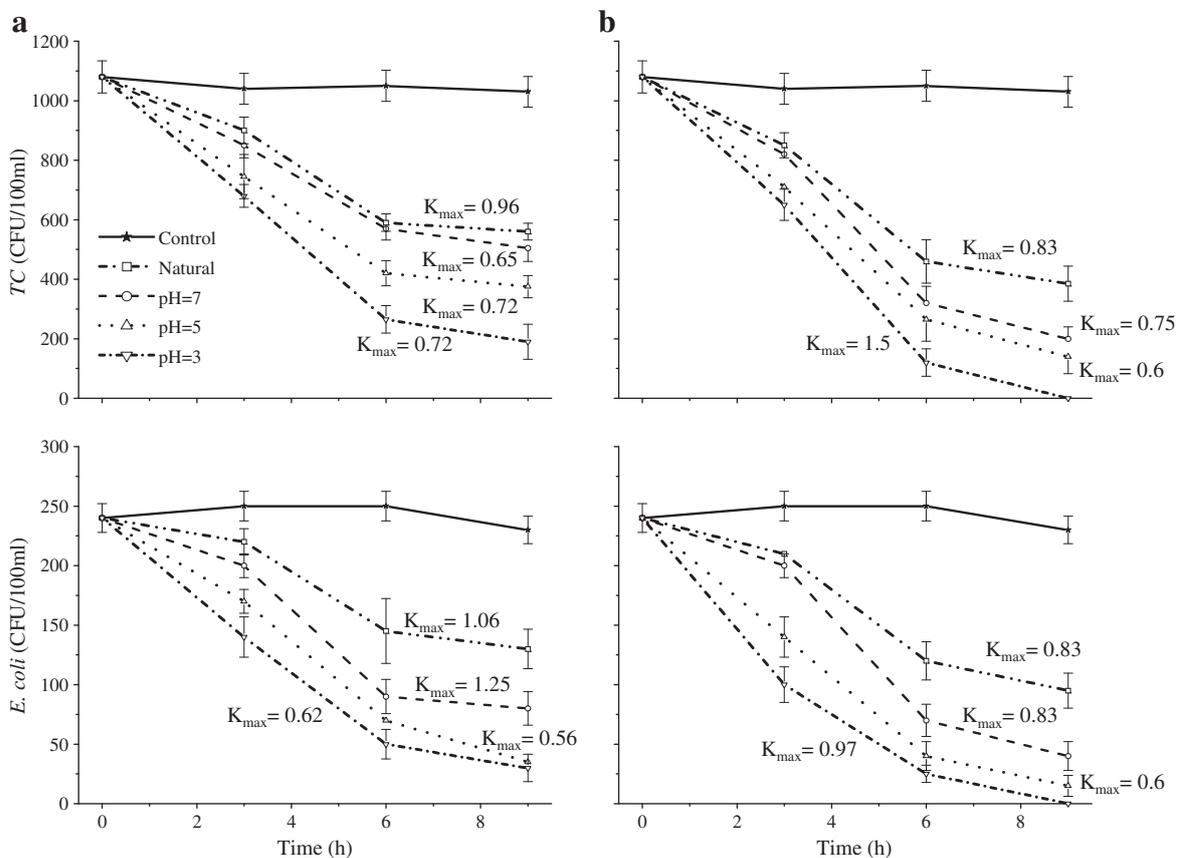


Fig. 4. TC and *E. coli* inactivation with different vinegar concentrations in; (a) SODIS, and (b) SOCO-DIS system.

the distribution system. The annual report of the National Health and Medical Research Council in 1996 (NHMRC, 1996) indicates that the consumption of food or beverages with low (2.5) or high pH (11) does not result in adverse health effects.

3.3.1. Disinfection by adding vinegar

Three different vinegar concentrations were used against three different adjusted initial pH values and the results of TC and *E. coli* inactivation under weak weather conditions are presented in Fig. 4. Each experiment was performed 3–5 times and the results presented are the mean average values for each point. The error bars in Figs. 4–8 show the minimum and maximum observed values for the respective microbial parameters.

The K_{max} (1/min) values shown in Figs. 4–7 were calculated based on the Geeraerd model, as described earlier, and represent the inactivation rate constants for TC and *E. coli*, with a corresponding coefficient of determination (R^2) of 1, except where mentioned. A complete application of the model, with tailing and shoulder, was observed for *E. coli* inactivation with SODIS at pH values of 8 (Natural sample) and 7. In most cases, however, modeling with tailing was unlikely for the observed data, due to the synergistic effects of vinegar with sunlight.

The comparisons were performed among three different vinegar concentrations and between the SODIS and SOCO-DIS systems. The results were compared with controlled and natural samples in each case, while pH was almost constant throughout the exposure time (results are not shown). There was an almost linear relationship between pH and disinfection efficiency with few exceptions i.e. a constant decrease in microbial concentrations was observed with a linear increase in pH value (Figs. 4–8). Disinfection was completed in terms of TC and *E. coli* inactivation in the SOCO-DIS system only at lowest pH value of around 3, corresponding to the final vinegar

concentration of approximately 4 mL per liter of rainwater. This high concentration of vinegar, around 0.4 percent by volume, may cause some odor or taste problems which were, however, overcome by using both lemon and vinegar in several combinations aiming at the same disinfection efficiency with low vinegar concentrations (Fig. 8). As shown in Fig. 4, disinfection efficiency increased by 40% by decreasing the initial pH values from almost 8 to nearly 3 in the SODIS and SOCO-DIS systems for a vinegar concentration of about 4 mL per liter of rainwater. This percent increase of the disinfection efficiency was evaluated by comparing the number of microbes at the end of 9 h exposure time in each case.

Low pH values may increase inactivation rates by presenting significant additional stress to the cells, for example by requiring the cells to expend energy maintaining pH homeostasis, thus accelerating the depletion of Adenosine triphosphate (ATP), the main energy storage and transfer molecule in the cells, and/or reducing equivalents. Biosynthetic reaction comes to halt as a consequence of ATP depletion and the cells lose their ability to maintain integrity, especially with respect to membrane systems [32]. The resulting metabolic stress due to low pH may also reduce the rate at which energy-consuming proteins in cells can scavenge reactive oxygen species (ROS), such as hydroxyl radical, superoxide radical anion, hydrogen peroxide, and singlet oxygen, and/or repair damaged Deoxyribonucleic Acid (DNA), thereby facilitating more rapid photoinactivation. ROS damage the external structures of microorganisms, such as the cell membranes.

3.3.2. Disinfection by adding lemon

As with the vinegar, three different lemon concentrations were used against three different adjusted initial pH values. However, the lemon concentrations were almost double that of the vinegar concentration for

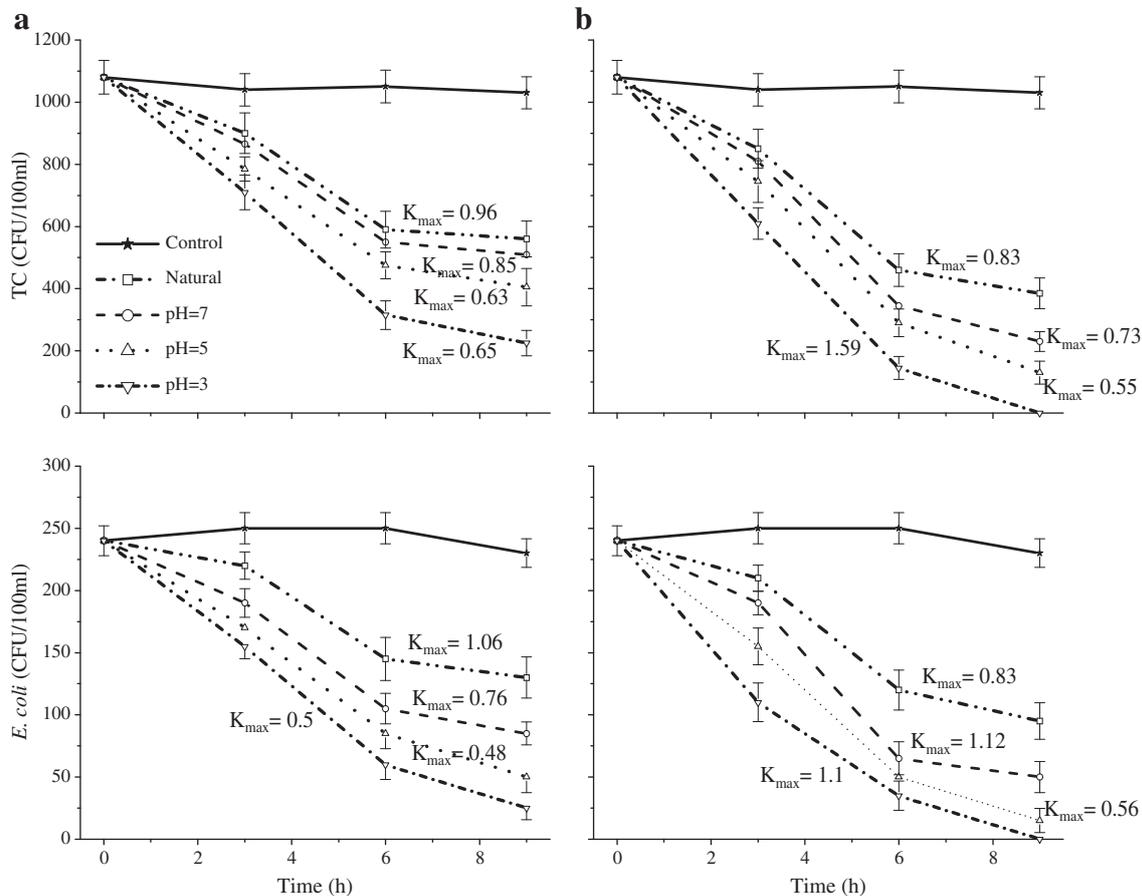


Fig. 5. TC and *E. coli* inactivation with different lemon concentrations in; (a) SODIS, and (b) SOCO-DIS system.

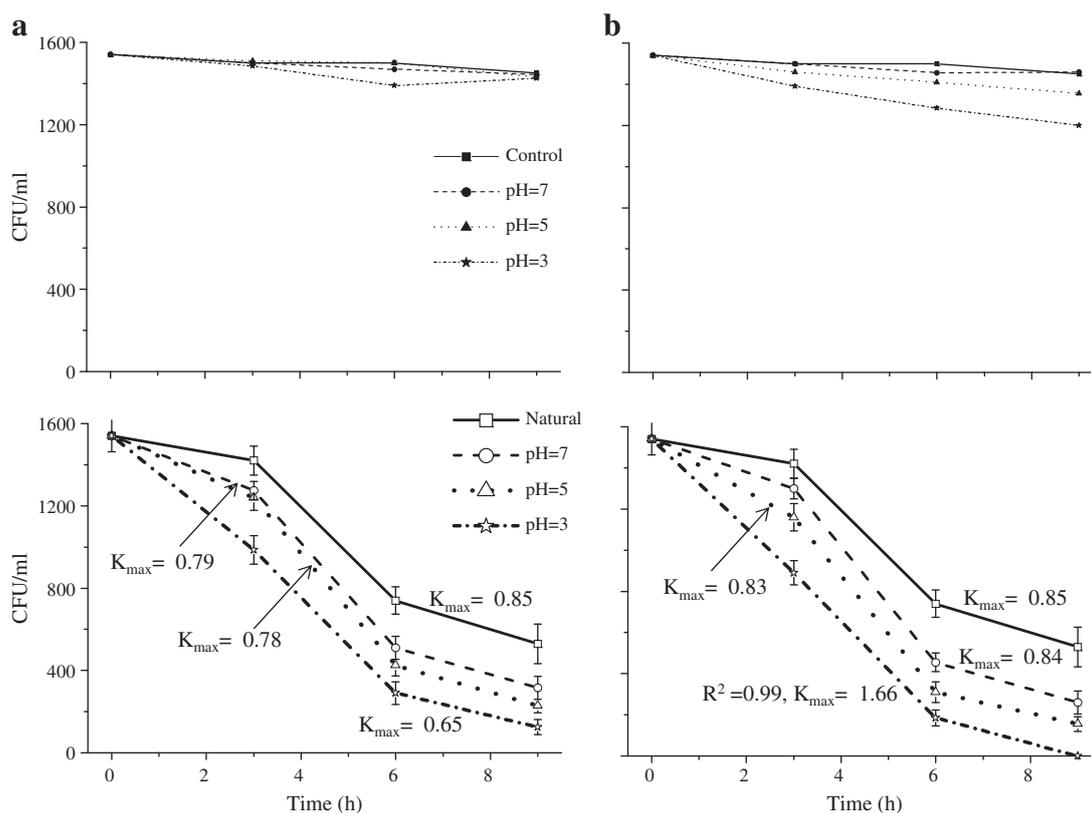


Fig. 6. HPC inactivation with different (a) lemon and (b) vinegar concentrations in SOCO-DIS system.

the respective pH values. The results of TC and *E. coli* inactivation under weak weather conditions are shown in Fig. 5.

The microbial inactivation efficiency was almost comparable when using the lemon as twice as the concentration of vinegar (comparing Figs. 4, 5 and Table 1). Disinfection efficiency increased by decreasing initial pH values and there was almost complete TC and *E. coli* removal in the SOCO-DIS system at lowest pH of around 3, while about 90% final inactivation was achieved for *E. coli* in SODIS by lowering the pH value to around 3. TC and *E. coli* inactivation increased by about 60% and 80%, respectively in SODIS at the lowest adjusted pH of 3 when compared to the sample without any lemon concentration. The synergetic effects of lemon with low pH in both SODIS and SOCO-DIS system accelerated the reaction process and, hence, enhanced the disinfection efficiency, as was the case with vinegar. The final lemon concentration for complete disinfection in terms of TC and *E. coli* was around 8 ml (0.8% by volume), which may also cause some taste problems.

Lemon, and lime juice concentrates possess intrinsic antimicrobial properties to eliminate *E. coli* and other bacterial pathogens in the event of postconcentration recontamination during the production of thermally concentrated fruit juices at high temperatures [33]. These bacterial pathogens, however, were recoverable from juice concentrates through 12 weeks of storage at -23°C only when samples were enriched in universal preenrichment broth for 72 h and plated on selective media [34]. In order to avoid the odor and taste problems and possible microbial re-growth due to the presence of nutrients in these food products, it is advisable to wash the used PET bottles on regular basis or to replace them with new ones since these are easily available.

3.3.3. Disinfection comparison between lemon and vinegar

The vinegar concentration required for the same pH and almost the same disinfection efficiency was almost half that of the corresponding lemon concentrations for TC and *E. coli* inactivation. In this section, a

quick comparison of lemon and vinegar is presented for HPC inactivation only in the SOCO-DIS system, as shown in Fig. 6.

The effects of lemon and vinegar on HPC inactivation were also evaluated without exposing samples to direct sunlight. Only vinegar has some effect on microbial inactivation and HPC was inactivated by about 20% by using around 0.4% by volume of vinegar, while HPC inactivation was around 7% in the case of lemon. One main difference between lemon and vinegar was the inactivation of HPC, which was not completed using a high test concentration of lemon while vinegar caused complete inactivation at the same pH value with almost half of the lemon concentration. Therefore, rainwater was not disinfected completely using lemon while, with vinegar, the problem of odor/taste may remain because of the 0.4% concentration of vinegar.

3.3.4. Combined effects of lemon and vinegar

Several combinations of lemon and vinegar were tried to obtain a pH of around 3 with the aim of attaining lemon and vinegar concentrations within the range of not causing any problems of taste or odor problems. The optimum combination for the best disinfection results was a lemon concentration of about 2.5 ml (0.25%) and a vinegar concentration of 1.7 ml (0.17%). The results for TC, *E. coli* and HPC inactivation under weak weather conditions only in the SOCO-DIS system are shown in Fig. 7. The K_{max} (1/min) values in Fig. 7 for TC, *E. coli* and HPC were calculated based on the Geeraerd model and represent the inactivation rate constants.

The use of the combined concentrations lowered the pH value from 8.7 to around 3.2. All three microbial parameters including TC, FC and *E. coli* were completely inactivated with this combination of lemon and vinegar as was the case when around 4 ml of vinegar alone was used (Fig. 4). HPC was almost disinfected completely, with a final concentration of around 15 MPN/ml against the drinking guideline value of 100 MPN/ml. Similar combinations can also be tried for complete disinfection under moderate weather conditions, with much less concentrations of vinegar or lemon, as the disinfection efficiency at this

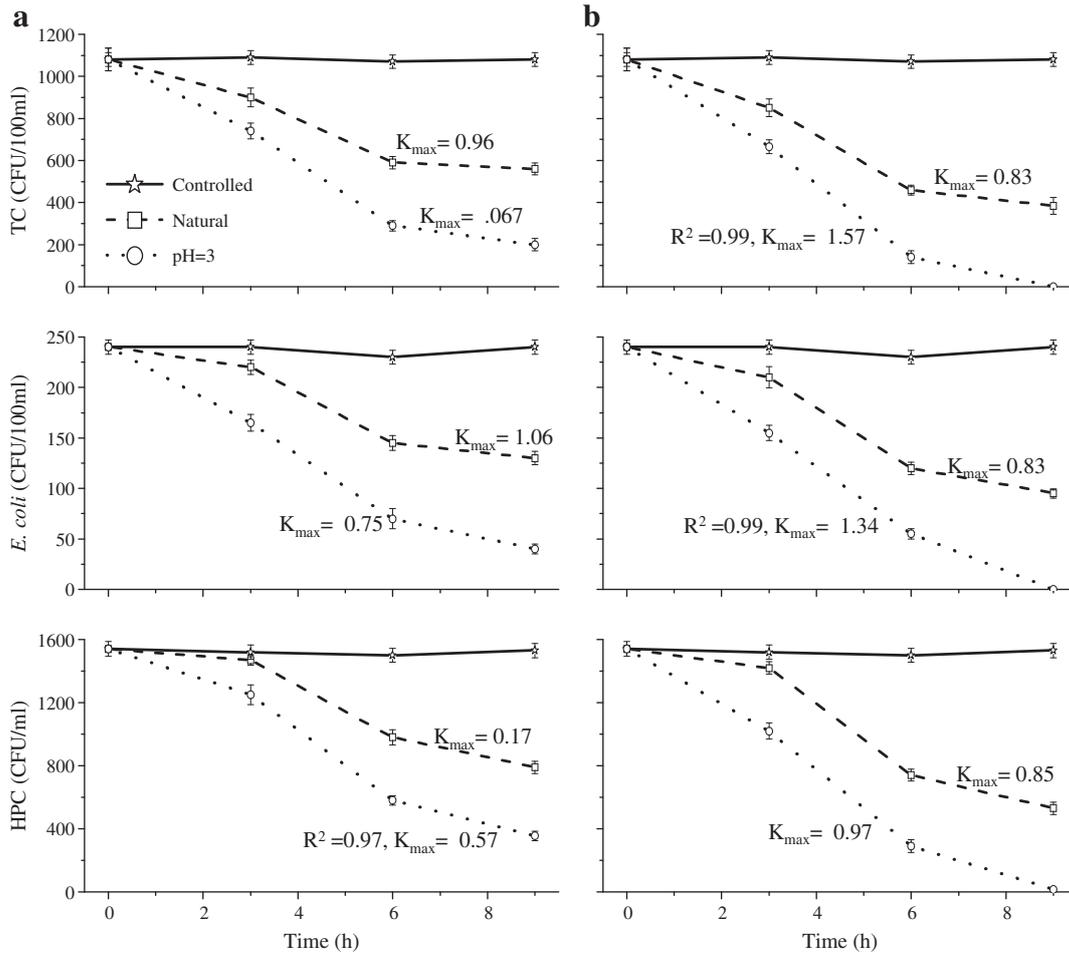


Fig. 7. Microbial inactivation with combined lemon and vinegar in (a) SODIS and (b) SOCO-DIS system.

stage is already around 80–90% for the SOCO-DIS system, without any modification of the natural conditions [26]. Finally, these concentrations of lemon and vinegar are low enough to produce an acceptable or even pleasant taste and ingestion of water containing such low concentrations should not pose health concerns, since stomach acid is far more acidic. However, acidic beverages may pose some risk of tooth decay.

Low pH alone is sufficient to explain the higher disinfection and a possible reason for the improved disinfection is the presence of acetic acid, present in vinegar, which resulted in decreased pH. This in turn can help to maintain high concentration of ROS, most probably due to the reduced scavenging rate of microorganisms. Another important factor could be the presence of natural organic matter (HPC concentrations) in the stored rainwater due to the catchment surfaces, which may also act as a photosensitizer and hence improve disinfection efficiency [35].

3.3.5. Comparison of lemon and vinegar for disinfection at moderate weather

E. coli inactivation comparison was performed between SODIS and the SOCO-DIS system by adding lemon and vinegar in both cases. A similar inactivation pattern for other microbial parameters (TC, FC and HPC) was observed (results not shown). This comparison was performed under moderate weather conditions to find the final concentrations of lemon and vinegar required for complete disinfection of all microbial parameters. All experiments were performed more than twice and the results are presented as the mean average values for each point in Fig. 8.

In the case of simple SODIS and the SOCO-DIS system, initial pH values were adjusted by adding diluted HCl (concentrations not measured), while the parent rainwater sample had a pH value of around 8.5. By comparing simple SODIS and SODIS with lemon and vinegar, it can be observed that none of the cases resulted in the complete inactivation of *E. coli*. Disinfection efficiency, however, increased by decreasing the pH, either by using HCl or lemon and vinegar. Inactivation was enhanced by about 30% by reducing the pH to only 7, from a natural pH value of 8.5, by using HCl. The addition of vinegar, however, increased inactivation by around 40% for the same pH adjustment. This trend of increasing inactivation by decreasing pH was not constant and finally about 10% better inactivation was observed after further lowering the pH from 7 to 3 when using vinegar compared with pH adjustment by lemon.

Almost similar trends were observed for the SOCO-DIS system, except that *E. coli* was completely inactivated at all pH values using vinegar and at pH 3 or even 5 when using lemon, and at pH 3 for the simple SOCO-DIS case where HCl was used for pH adjustment. By comparing Fig. 8 (parts b, d and f), it is obvious that vinegar is more effective than lemon or HCl for microbial removal for the same pH value, which explains the importance of choosing the proper catalyst (food product) for enhancing the disinfection.

4. Conclusions

The application of solar-based disinfection technologies to obtain potable stored rainwater to meet the daily demands of individuals or a family in rural/semi urban areas of developing countries is

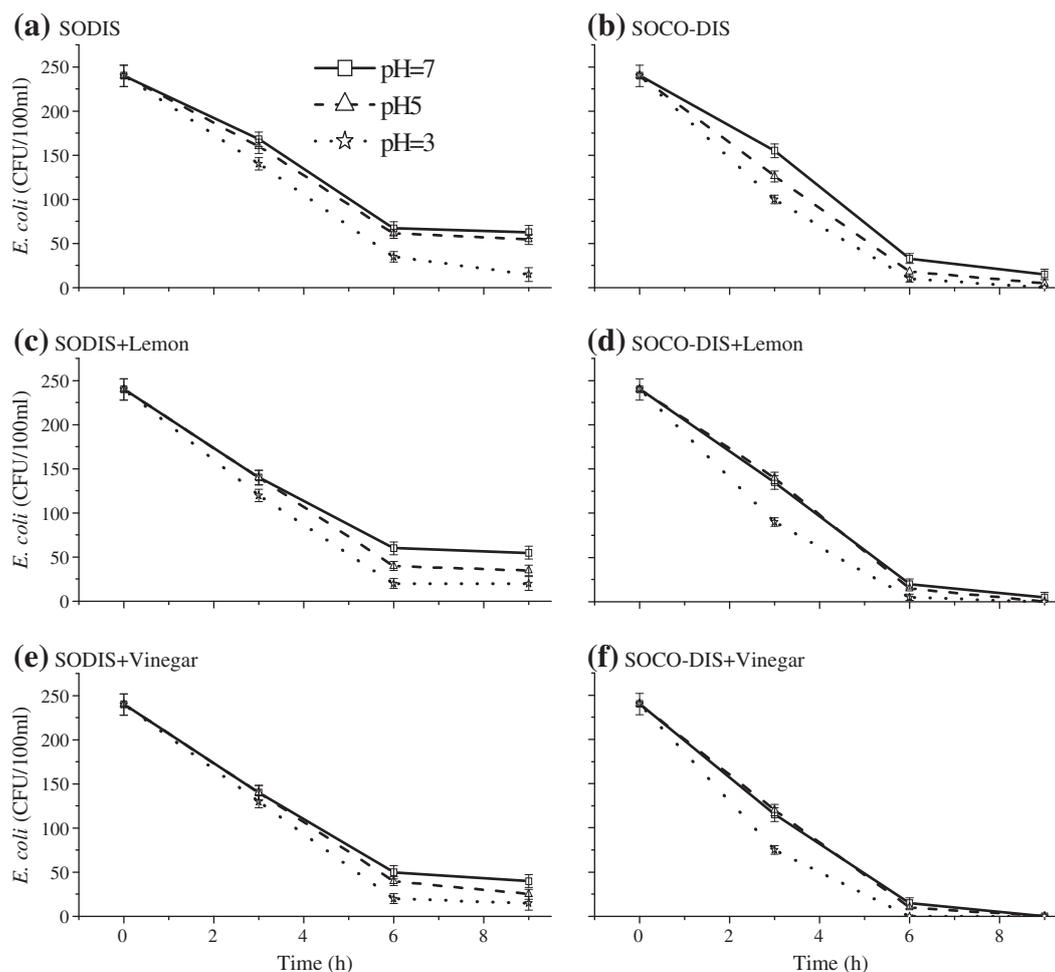


Fig. 8. *E. coli* inactivation in SODIS and SOCO-DIS and with lemon and vinegar.

investigated by adding some commonly available and inexpensive food preservatives to act as catalysts. For this purpose, lemon and vinegar were used to enhance the efficiency of SOCO-DIS system, based on the better performance of solar-based disinfection systems at low pH values [14,26] for complete disinfection of rainwater, especially under weak weather conditions.

Lemon and vinegar both increased the disinfection efficiency by around 40% in SODIS, however, complete inactivation was not observed. In SOCO-DIS system, on the other hand, completely disinfected rainwater was obtained under weak weather conditions, except for the HPC inactivation when using lemon. The amount of vinegar required for the same disinfection efficiency was almost half that of the corresponding amount of lemon, which highlights the proper selection and choice of catalyst for disinfecting rainwater using sunlight.

To avoid any taste or odor problems due to high concentrations of lemon or vinegar, optimum combinations of both food products were tried and satisfactory results were obtained at the lowest pH values of around 3, without any of the problems of taste or odor associated with high concentrations of lemon or vinegar alone. The combination of 2.5 ml (0.25%) of lemon and 1.7 ml (0.17%) of vinegar produced the same results as obtained by using 7.5 ml (0.75% by volume) of lemon or 3.8 ml (0.38% by volume) of vinegar, separately.

Practical benefits of these additions include the potential and possibly considerable application of solar based disinfection systems for the rapid disinfection of stored rainwater under weak sunlight conditions and for routine disinfection in larger and more practical containers for complete disinfection in small scale potable water supply systems at the community level under moderate to strong weather conditions.

Acknowledgements

This work was supported by Gyeonggi Sea Grant Program funded by the Ministry of Land, Transport and Maritime Affairs of Korean government, SNU SIR Group of the BK21 research Program funded by the Ministry of Education, Science and Technology and Research Institute of Engineering Science, Seoul National University.

References

- [1] M.T. Amin, M.Y. Han, Water environmental and sanitation status in disaster relief of Pakistan's 2005 earthquake, *Desalination* 248 (2009) 436–445.
- [2] M.B. Ibrahim, Rainwater harvesting for urban areas: a success story from Gadarif City in Central Sudan, *Water Resour. Manag.* 23 (2009) 2727–2736.
- [3] L. Handia, J.M. Tembo, C. Mwiindwa, Potential of water harvesting in urban Zambia, *Phys. Chem. Earth* 28 (2003) 893–896.
- [4] H.C. Preul, Rainfall-runoff water harvesting prospects for greater Amman and Jordan, *Water Int.* 19 (2) (1994) 82–85.
- [5] T. Thomas, Domestic water supply using rainwater harvesting, *Build. Res. Inf.* 26 (2) (1998) 94–101.
- [6] A.A. Murad, H. Al-Nuaimi, M. Al-Hammadi, Comprehensive assessment of water resources in the United Arab Emirates (UEA), *Water Resour. Manag.* 21 (2007) 1449–1460.
- [7] M. Ruth, C. Bernier, N. Jollands, N. Golubiewski, Adaptation of urban water supply infrastructure to impacts from climate and socioeconomic changes: the case of Hamilton, New Zealand, *Water Resour. Manag.* 21 (2007) 1031–1045.
- [8] E. Wheida, R. Verhoeven, An alternative solution of the water shortage problem in Libya, *Water Resour. Manag.* 21 (2007) 961–982.
- [9] J.K. O'Hara, P. Georgakakos, Quantify the urban water supply impacts of climate change, *Water Resour. Manag.* 22 (2008) 1477–1497.
- [10] T. Clasen, D. Thao, S. Boisson, O. Shipin, Microbiological effectiveness and cost of boiling to disinfect water in rural Vietnam, *Environ. Sci. Technol.* 42 (12) (2008) 4255–4260.

- [11] M.D. Sobsey, C.E. Stauber, L.M. Casanova, J.M. Brown, M.A. Elliott, Point of use household drinking water filtration: a practical, effective solution for providing sustained access to safe drinking water in the developing world, *Environ. Sci. Technol.* 42 (12) (2008) 4261–4267.
- [12] R.M. Conroy, M.E. Meegan, T. Joyce, K.G. McGuigan, J. Barnes, Solar disinfection of drinking water and diarrhoea in Maasai children: a controlled field trial, *Lancet* 348 (9043) (1996) 1695–1697.
- [13] A. Rose, S. Roy, V. Abraham, G. Holmgren, K. George, V. Balraj, S. Abraham, J. Muliylil, A. Joseph, G. Kang, Solar disinfection of water for diarrhoeal prevention in southern India, *Arch. Dis. Child.* 91 (2) (2006) 139–141.
- [14] M.T. Amin, M.Y. Han, Roof-harvested rainwater for potable purposes: application of solar disinfection (SODIS) and limitations, *Water Sci. Technol.* 60 (2) (2009) 419–431.
- [15] M. Wegelin, S. Canonica, A.C. Alder, D. Marazuela, M.J.-F. Suter, Th.D. Bucheli, O.P. Haefliger, R. Zenobi, K.G. McGuigan, M.T. Kelly, P. Ibrahim, M. Larroque, Does sunlight change the material and content of polyethylene terephthalate (PET) bottles? *J. Water Supply Res. T.* 50 (3) (2001) 125–133.
- [16] B. Sommer, A. Marino, Y. Solarte, M.L. Salas, C. Dierolf, C. Valiente, D. Mora, R. Rechsteiner, P. Setter, W. Wirojanagud, H. Ajarmeh, A. Al-Hassan, M. Wegelin, SODIS—an emerging water treatment process, *J. Water Supply Res. T.* 46 (1997) 127–137.
- [17] S.C. Kehoe, T.M. Joyce, P. Ibrahim, J.B. Gillespie, R.A. Shahar, K.G. McGuigan, Effect of agitation, turbidity, aluminium foil reflectors and container volume on the inactivation efficiency of batch-process solar disinfectors, *Water Res.* 35 (2001) 1061–1065.
- [18] S. Gelover, A.G. Luis, K. Reyes, M.T. Leal, A practical demonstration of water disinfection using TiO₂ films and sunlight, *Water Res.* 40 (2006) 3274–3280.
- [19] R.J. Watts, S. Kong, M.P. Orr, G.C. Miller, B.E. Henry, Photocatalytic inactivation of coliform bacteria and viruses in secondary wastewater effluent, *Water Res.* 29 (1995) 95–100.
- [20] P. Zhang, R. Scrodato, G. Germano, Solar catalytic inactivation of *Escherichia coli* in aqueous solution using TiO₂ as catalyst, *Chemosphere* 28 (1994) 607–611.
- [21] D.M. Blake, P.-C. Maness, Z. Huang, E.J. Wolfrum, J. Huang, W.A. Jacoby, Application of the photocatalytic chemistry of titanium dioxide to disinfection and the killing of cancer cells, *Separ. Purif. Method* 28 (1999) 1–50.
- [22] D.D. Sun, J.H. Tay, K.M. Tan, Photocatalytic degradation of *E. coli* form in water, *Water Res.* 37 (2003) 3452–3462.
- [23] F.M. Salih, Enhancement of solar inactivation of *Escherichia coli* by titanium dioxide photocatalytic oxidation, *J. Appl. Microbiol.* 92 (2002) 920–926.
- [24] E.F. Duffy, F. Al-Touati, S.C. Kehoe, O.A. McLoughlin, L. Gill, W. Gernjak, I. Oller, M.I. Maldonado, S. Malato, J. Cassidy, R.H. Reed, K.G. McGuigan, A novel TiO₂-assisted solar photocatalytic batch-process disinfection reactor for the treatment of biological and chemical contaminants in domestic drinking water in developing countries, *Sol. Energy* 77 (2004) 649–655.
- [25] M.B. Fisher, C.R. Keenan, K.L. Nelson, B.M. Voelker, Speeding up solar disinfection (SODIS): effects of hydrogen peroxide, temperature, pH, and copper plus ascorbate on the photoinactivation of *E. coli*, *J. Water Health* 60 (1) (2008) 35–51.
- [26] M.T. Amin, M.Y. Han, Roof-harvested rainwater for potable purposes: application of solar collector disinfection (SOCO-DIS), *Water Res.* 43 (2009) 5225–5235.
- [27] APHA, Standard Methods for the Examination of Water and Wastewater, 20th ed. American Public Health Association, Washington, DC, 1999.
- [28] R.H. Reed, Solar inactivation of faecal bacteria in water: the critical role of oxygen, *Lett. Appl. Microbiol.* 24 (1997) 276–280.
- [29] A.H. Geeraerd, V.P. Valdramidis, J.F. van Impe, GlnaFit, a freeware tool to assess non-log-linear microbial survivor curves, *Int. J. Food Microbiol.* 10 (2005) 95–105.
- [30] A.H. Geeraerd, C.H. Herremans, J.F. van Impe, Structural model requirements to describe microbial inactivation during a mild heat treatment, *Int. J. Food Microbiol.* 59 (3) (2000) 185–209.
- [31] M. Wegelin, S. Canonica, K. Mechsner, T. Fleischmann, F. Pesaro, A. Metzler, Solar water disinfection: scope of the process and analysis of radiation experiments, *J. Water Supply Res. T.* 43 (1994) 154–169.
- [32] E.A. Foegeding, T.C. Lanier, H.O. Hultin, Owen R. Fennema, Characteristics of edible muscle tissues, *Food Chemistry*, Vol. 3, 1996, pp. 879–942, Chapter 15.
- [33] M.C.L. Nogueira, O.A. Oyarza^a Bal, D.E. Gombas, Inactivation of *Escherichia coli* O157:H7, *Listeria monocytogenes*, and *Salmonella* in Cranberry, Lemon, and Lime Juice Concentrates, *J. Food Prot.* 66 (9) (2003) 1637–1641.
- [34] O.A. Oyarza^a Bal, M.C.L. Nogueira, D.E. Gombas, Survival of *Escherichia coli* O157:H7, *Listeria monocytogenes*, and *Salmonella* in Juice Concentrates, *J. Food Prot.* 66 (9) (2003) 1595–1598.
- [35] T.P. Curtis, D.D. Mara, S.S. Silva, Influence of pH, oxygen, and humic substances on ability of sunlight to damage fecal coliforms in waste stabilization pond water, *Appl. Environ. Microbiol.* 58 (1992) 1335–1343.