Demonstration and evaluation of germicidal UV-LEDs for point-of-use water disinfection

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ABSTRACT

Ultraviolet (UV) irradiation is a common disinfection option for water treatment in the developed world. There are a few systems installed in developing countries for point-of-use treatment, but the low-pressure mercury lamps currently used as the UV irradiation source have a number of sustainability issues including a fragile envelope, a short lifetime of approximately one year, and they contain toxic mercury. UV light emitting diodes (LEDs) may present solutions to many of the sustainability issues presented by current UV systems. LEDs are small, efficient, have long lifetimes, and do not contain mercury. Germicidal UV LEDs emitting at 265 nm were evaluated for inactivation of *E. coli* in water and compared to conventional low-pressure UV lamps. Both systems provided an equivalent level of treatment. A UV-LED prototype was developed and evaluated as a proof-of-concept of this technology for a point-of-use disinfection option, and the economics of UV-LEDs were evaluated.

Key words | household water treatment, light emitting diodes, ultraviolet light

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INTRODUCTION

Diarrheal illnesses are one of the leading causes of morbidity and mortality in developing countries (Pruss 2002; WHO 2002). In many analyses of interventions to reduce diarrhea, "improved water quality" is shown to have a lower effect than other interventions such as sanitation and hygiene. However, these reviews focus upon source water quality improvements rather than improvements at point-of-use (Gundry *et al.* 2004). Fewtrell & Colford (2004), showed the increased impact of treating water at the household level compared to treating at the source. This information has created an interest in household water treatment technologies. A number of point-of-use technologies have been evaluated including boiling, biosand filtration, chlorination, chlorination plus flocculation, solar disinfection (SODIS), and ceramic filters (Sobsey *et al.* 2008). Disinfection using ultraviolet (UV) radiation in the UV-C range may be a more favorable option for many applications. It does not utilize chemicals and disinfects at much higher rates than SODIS which utilizes temperature and radiation in the UV-A range.

UV disinfection is a well-established disinfection technology that has been used in centralized water and wastewater facilities in developed countries for decades. UV radiation inactivates bacteria, viruses, and protozoa, with the benefits of no taste and odor issues, no known disinfection byproducts (DBPs), no danger of overdosing, relatively fast treatment rates compared to sand and ceramic filters, and low-maintenance requirements. Over the last ten years, small UV systems have become available, including commercially available household systems and the low-cost, locally manufactured UV-Tube system that have become an appropriate treatment option for developing communities in a number of countries including Mexico, Sri Lanka, and India (Brownell *et al.* 2008).

For developing communities, UV disinfection can be an improvement over other treatment options, such as

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chemical disinfection, for many applications, but there are sustainability issues that arise from current low-pressure lamp systems in use. They use toxic mercury as the UV radiation source and typically only last for 8,000–10,000 hours, at which time communities are faced with a number of issues: finding and paying for replacement lamps, transporting these fragile glass and filament tubes, and disposing of mercury contained in the used lamp in areas that do not typically have a toxic waste disposal system (US EPA 2006).

UV light emitting diodes (LEDs) may provide solutions to many of the sustainability issues of UV mercury lamps. They are small (5–9 mm diameter), and do not contain glass, filament or mercury, aiding their transport and disposal (Bettles *et al.* 2007). Warm-up time is not required for LEDs, saving energy and allowing for intermittent use and quick recovery from a power failure–important characteristics for rural applications especially. LEDs are replacing a number of light sources currently utilized today including traffic lights and household lights. LEDs have an excellent track record for lowering system costs through energy savings, lower maintenance, and longer replacement intervals. The average electrical-to-germicidal efficiency of low-pressure UV mercury tube lamps is 35–38% (US EPA 2006). Visible LEDs can operate at 75% efficiency for ten years (100,000 hours) (Bettles *et al.* 2007). Currently, the efficiencies of UV-LEDs are less than 1% with lifetimes of around 1,000 hours (Bettles *et al.* 2007; Gaska 2007). Although research of this technology is still in its infancy, improvements to UV-LEDs are expected to occur rapidly following visible LED source trajectories, resulting in a high efficiency, low power input.

The availability of specific output wavelengths using UV-LEDs may also increase their inactivation efficacy. UV-LEDs currently operate in the wavelength range of 247–365 nm (Gaska 2007). Effective UV sources should emit high intensities in the peak absorbance wavelengths of DNA–the germicidal target of UV photons. However, germicidal effectiveness as a function of wavelength can vary for different microorganisms and may differ from the DNA absorbance spectrum. Supplementing peak DNA wavelengths with other UV emissions may provide a synergistic disinfection effect, increasing the effectiveness of UV inactivation of pathogens (Mamane-Gravetz *et al.* 2005; US EPA 2006; Linden *et al.* 2007). Low-pressure lamps are monochromatic (254 nm) and some pathogens, such as adenovirus, are not most effectively inactivated at this wavelength. Medium Pressure lamps are polychromatic, but peak intensities occur at set wavelengths based on the emission properties of mercury. A distinct advantage over conventional UV sources is that UV-LED systems can incorporate an LED array of differing UV wavelengths, maximizing their combined germicidal effect. This would allow units to be custom designed based on the specific pathogens of concern in source waters, or for a broad range of pathogens under a single system.

Limited research has been conducted on the effectiveness of UV-LEDs for water disinfection. Most of the data available are for LEDs that emit light in the UVA range (320–400 nm), which is less efficient at disinfection than light in the germicidal range of UVC (200–280 nm) since it is poorly absorbed by DNA (Sinha & Häder 2002; ISO 21348 2007). UVA radiation inactivates microorganisms by damaging proteins and producing hydroxyl and oxygen radicals that can destroy cell membranes and other cellular components (Sinha & Häder 2002). This process takes more time than the damage produced by UV-C, which directly effects the DNA of microorganisms by producing cyclobutane thymine dimers, among other products, inactivating them without intermediate steps (Grossweiner & Smith 1989). Hamamoto *et al.* (2007) demonstrated the ability of UVA-LEDs at 365 nm to inactivate bacteria in water. They found that *E. coli* DH5α were reduced by > 5 log at a dose of 315 J/cm^2 approximately 30,000 times higher dose than required for UV 254 nm. Sandia National Laboratories documented inactivation of *E. coli* with LEDs in the UVC range at 270 nm (Crawford *et al.* 2005), and found comparable inactivation to LP UV. Sensor Electronics Technologies (SET) has also demonstrated inactivation of *E. coli* B using 265–310 nm UV-LEDs (Gaska 2007), reporting wavelength dependent inactivation with inactivation decreasing by more than 6 orders of magnitude from 265 nm to 310 nm.

Research objectives

The goal of this research was to evaluate the efficacy of Ultraviolet Light Emitting Diode (UV-LED) technology for

the development of point-of-use (POU) water disinfection systems to improve public health in rural communities in a sustainable, environmentally responsible manner. There are a number of POU technologies available, but the application of UV-LEDs as a disinfection source will provide an additional technology to the POU toolbox that will enable longer-life disinfection systems with low user input and very low energy cost compared to current low-pressure mercury lamps. This will improve public health by increasing system reliability and decreasing maintenance needs.

Specifically, this research evaluated the use of UV-LEDs at 265 nm for inactivation of *E. coli* in water through the following objectives: (1) Compare the inactivation efficiency of UV-LEDs at 265 nm to conventional low-pressure lamps (254 nm) for inactivation of *E. coli*, (2) Evaluate a point-of-use UV-LED flow-through prototype, and (3) Determine if UV-LEDs are a feasible option for water treatment based on economics and current state of the technology.

METHODS

Microbial methods

E. coli K12 (ATCC #29425) was used as an indicator organism to compare the efficiency of the LP and UV-LED systems, and to evaluate the UV-LED prototype. One colony (to assure genetic homogeneity) was obtained from a tryptic soy agar (TSA, Difco #236950) plate after 24 hours of incubation at 37°C and added to 10 mL of sterile tryptic soy broth (TSB, Cellgro #61-412-RO) in a sterile 15 mL vial. The vial was rapidly vortexed to break up the colony and then the 10 mL solution was added to 90 mL of TSB in a sterile 250 mL glass bottle with a sterile magnetic stir bar. The stock solution was incubated at 37°C on a stir-plate to assure constant mixing and oxygen levels throughout the stock. This solution was kept at 4°C for less than 2 weeks to inoculate future stock solutions. Purity was verified by streak plating and visual observation on TSA.

A growth curve was developed based on the optical density at 600 nm (OD600), measured every 30 minutes, and cultured colonies to identify the log growth phase. Tests were conducted at log growth phase. The *E. coli* were washed three times in phosphate buffer solution (PBS) by centrifuging and added to 194 mL of PBS to achieve a concentration of approximately 10^6 CFU *E. coli* per mL for batch irradiation testing. The ultraviolet absorbance was adjusted for flow-through tests by varying the *E. coli* preparation steps (washing).

After irradiation, each sample was successively vortexed and serially diluted. *E. coli* concentrations were measured using the spot plating method (Gaudy *et al.* 1962), which is advantageous because four dilutions of five replicates each can be read on one 100 mm diameter plate, which would otherwise require twenty 60 mm diameter plates using the vacuum filtration method. Once the spots had completely dried, the plates were placed upside down in a 37°C incubator and incubated for 24 hours before colonies were counted. Spots with 3 to 30 colonies were recorded (CFU/0.01 mL). If two dilutions had results that fell into this range, the lower dilution (more colonies in each spot) was chosen.

Experimental set-up

Low-pressure (LP) UV lamps were housed in a UV collimated beam apparatus (Bolton & Linden 2003). A UV-LED batch irradiation system was designed with an array of three UV-LEDs using a circuit wire-wrapped with 30 gauge wire, to an electronic Perfboard (a fiberglass board with holes every 2.5 mm). A 150-ohm resistor was wired in series with each LED to create 6 volts across each LED at 20 amps with a 9 volt input voltage from a power supply. These values were within manufacturer specifications for voltage and current. Socket pins were wire-wrapped to the Perfboard to hold the LEDs in place for easy removal and replacement. A flow-through prototype consisting of a compact row of ten UV-LEDs was created using similar electronics to the batch system. The LEDs were placed over a 6.5 mm × 6.5 mm aluminum channel 1 mm above the water surface, with a water depth of 7 mm. The 265 nm hemispherical UV LEDs were purchased from Sensor Electronic Technology, Inc (SET) (Columbia, South Carolina).

Irradiance measurements

Irradiance for the LP and LED light sources was measured with a radiometer (International Light IL1400A, SEL

240/TD detector) calibrated at 254 nm. The manufacturer's detector response curve was used to adjust the radiometer reading to the 265 nm peak output wavelength of the LEDs. The radiometer measurements were checked and corroborated using an iodide/iodate actinometer (Rahn *et al.* 2003). The absolute irradiance and spectral output of each LED was also evaluated using an Ocean Optics spectrometer (USB 2000 + , Dunadin, FL).

UV irradiation

All tests were completed within two hours and irradiated samples were covered to minimize photoreactivation as much as possible.

To benchmark the efficiency of LP UV, the collimated beam apparatus was used to expose 40 mL portions of *E. coli* spiked PBS at UV fluences ranging from 0 to 20 mJ/cm^2 in a sterile 50 mL glass crystallization dish (2.2 cm diameter) stirred with a sterile magnetic stir bar. The UV-LED devices were evaluated by exposing 7 mL of *E. coli* spiked PBS in a 10 mL beaker (2.2 cm diameter) stirred with a sterile magnetic stir bar to UV doses between 0 and 20 mJ/cm^2.

The UV-LED prototype was evaluated by flowing *E. coli* spiked PBS and *E. coli* spiked natural water (collected from a local pond) through the system. Initial *E. coli* concentration was tested by running the sample through the prototype with the LEDs turned off. Log reduction of *E. coli* was evaluated for multiple flow rates and multiple UV absorbance values. The system was disinfected between tests using a low concentration chlorine solution.

UV dose calculations

The average irradiance in the UV-exposed sample was calculated according to Bolton and Linden (Bolton & Linden 2003). A petri factor of 0.98 and 1 were determined for the LP and LED systems, respectively, and a reflection factor for water of 0.975 was used. The water factor accounted for the UV absorbance of the water through the sample water depth at 254 nm and 266 nm for the LP and LED systems, respectively (measured with a spectrophotometer, HACH DR 5000).

Irradiation time was controlled by a manual shutter for LP tests and by turning on/off the LEDs. The LP lamps and LEDs were allowed to warm-up for 10 minutes before tests and the LEDs were turned off for a maximum of 10 seconds while tests were being set-up, which did not significantly affect the irradiance upon turning back on.

The *E. coli* colonies were averaged for the five spots and converted to CFU/mL by multiplying by the dilution factor. The log reduction ($\log N_0/N$) was calculated for each dose based on the initial non-irradiated *E. coli* concentration, N_0, (CFU/mL) and the concentration of *E. coli* post-irradiation, N (CFU/mL).

RESULTS AND DISCUSSION

Irradiance during warm-up time

The irradiance over time (0, 1, 2, 5, 10, and 20 minutes from start) was measured to determine the warm-up time of both the low-pressure (LP) lamps and the UV-LEDs. Over the first 10 minutes after start-up, the irradiance of the UV-LEDs decreases by about 7% and the irradiance of the LP lamps increases by about 20%, after which time both sources level out (Figure 1).

Seven LEDs from SET were tested including four flat top 265 nm LEDs, one flat top 250 nm LED, one flat top 280 nm LED, and one hemispherical lens 280 nm LED. The spectral irradiance of each LED was measured with a spectrometer (Ocean Optics USB 2000 +) as presented in Figure 2. The UV-LEDs from SET had a broader bandwidth than the monochromatic 253.7 nm produced by low-pressure lamps. The full width at half maximum (FWHM), measured across the spectral output at 50% of the peak

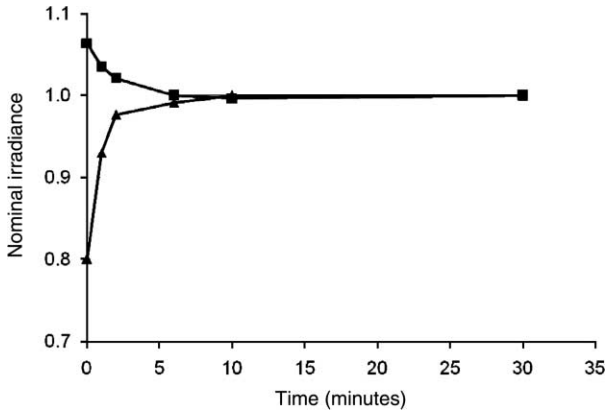

Figure 1 | Warm-up time for UV-LEDs (■) versus Low Pressure Lamps (▲).

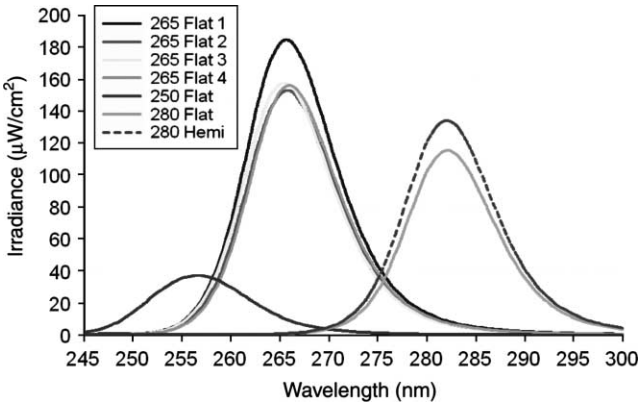

Figure 2 | Irradiance and spectral emission from 250, 265, and 280 nm UV-LEDs.

irradiance, was 11 nm for the 265 nm LED, slightly lower than the manufacturer FWHM specification of 12 nm. The broader emission spectra could have implications for system designs, paricularly if very specific wavelengths are desired.

Irradiance from a single 265 nm hemispherical lens UV-LEDs was measured at distances from the source up to four cm, to estimate the effect of distance on irradiance changes, specifically for future modeling and estimating UV dose for the prototype unit. The irradiance output from the 265 nm LEDs tested varied from 60 to 30 $\mu W/cm^2$, at distances of 0.5 to 4 cm as illustrated in Figure 3.

Inactivation of *E. coli*

Low-pressure versus UV-LEDs

Log reduction of *E. coli* K12 appears to be slightly improved for the LED source at low doses and approximately the same at higher doses, based on twenty three and eighteen data points in duplicate for the LED and LP sources respectively, as illustrated in Figure 4.

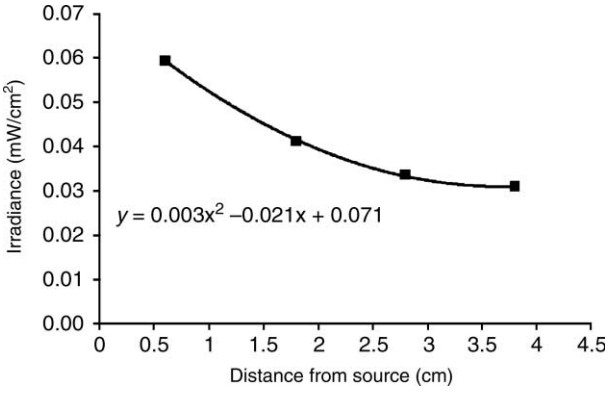

Figure 3 | Irradiance of one 265 nm LED as a function of distance from the LED.

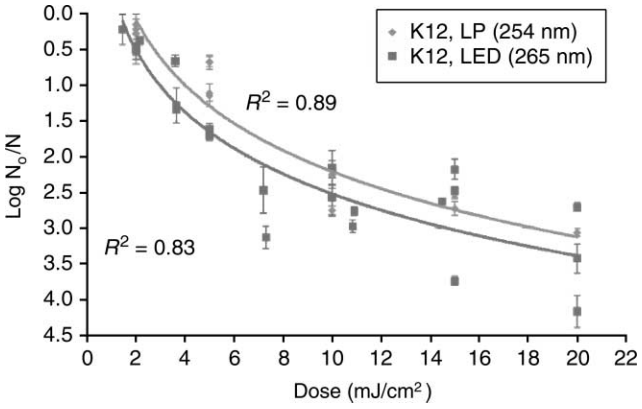

Figure 4 | Log reduction of *E. coli* K12 by irradiation from low-pressure lamps (254 nm) and LEDs (265 nm). Data lines represent logarithmic regression and error bars represent one standard deviation of the mean of quintuplicates.

The relationship of log inactivation versus dose received was modeled using a logarithmic regression. Based on results of paired *t*-tests conducted over the log inactivation data at each dose (2, 5, 10, 15, and 20 mJ/cm^2) for low-pressure versus LED sources, it can not be concluded that the low-pressure and LED sources are statistically different for the inactivation of *E. coli* K12 at a 95% confidence, although there is a statistically significant difference at a 90% confidence level.

UV-LED flow-through prototype

The ten-LED prototype was evaluated using biodosimetry with *E. coli* K12. The linear trendlines for log reduction with varying UV absorbance values all have a similar slope (within one log reduction per one liter per hour) and the waters with lower UV absorbance values are disinfected to the same level as waters with higher UV absorbance values when lower flow rates were used (Figure 5). The inactivation in natural water (UV absorbance of 0.259) was similar to the PBS samples.

In order to compare the prototype to commercial systems, the dose provided for a given flow rate in mL/min and influent water UV transmittance (UVT) are needed. The log reduction of *E. coli* for a given UV-LED dose was calculated based on the logarithmic regression model (log reduction = 1.25 × Ln(Dose) − 0.3665) on the

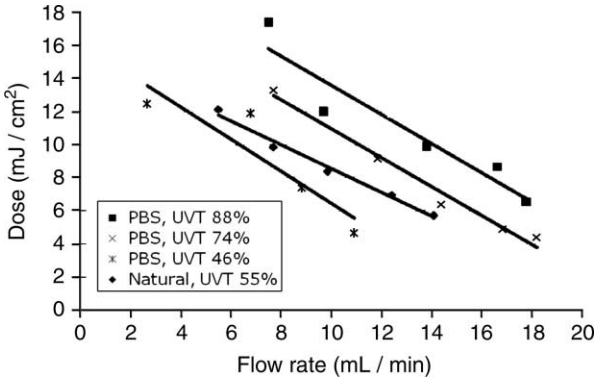

Figure 5 | Dose for given flow rate and UV transmittance for *E. coli* K12 spiked into PBS and natural water.

E. coli inactivation data presented in Figure 4. For a UVT of 88%, a dose of 10 mJ/cm^2 can be achieved at a flow rate of 14 mL/min. For a UVT of 74%, the flow rate required for a dose of 10 mJ/cm^2 is 11.1 mL/min. Therefore, forty LEDs (four rows of 10 LEDs in the same geometry as the prototype) at a flow rate of 11.1 mL/min could provide a dose of roughly 40 mJ/cm^2, a typical dose for commercial UV disinfection systems. Forty LEDs emitting at 265 nm have a total rated output of 14.4 mW of power, therefore 1.3 mW are needed per mL/min flow rate to achieve a dose of 40 mJ/cm^2. This value is used to compare LED and LP systems in Table 1. However, direct comparison is highly dependent on reactor hydraulics and the comparison presented is used only as a case study based on the LED prototype evaluated. The power requirements for UV-LED systems have the potential to be reduced by optimizing reactor geometry and water depth. The use of UV-LEDs may facilitate optimization through creative system design options based on their small individual size.

Table 1 | Comparison of current and projected future UV-LEDs with LP systems

			UV-LEDs	
	UV-Tube	Sterilight	Base case	3–4 year projection
MW/Lamp (output)	15,000	10,000	0.36	100
Lifetime (hrs)	9,000	9,000	1,000	10,000
Cost ($/mW)*	0.0013	0.0055	664	0.1
Flow rate (mL/min)	6,000	1,890	55.55	55.55
Total mW (output)†			72.22	72.22
Number of LEDs			201	1
Upfront lamp cost ($)	20	55	47,943	7
3 year cost	60	165	1,260,063	21
20 year cost	389	1,071	8,400,421	123

*$ values in USD. Current LED cost based on purchase price from SET December 2007.
†Total power output was calculated by multiplying the flow rate by 1.3 as found in the prototype testing.

Evaluation of current and future UV-LED technology

LEDs that emit light in the germicidal wavelength range are a relatively new technology and current values for cost, output power, and lifetime do not at present allow them to be a viable option for the replacement of low-pressure lamps used for drinking water disinfection, especially in developing communities. Based on a household system that needs to provide twenty liters per person per day for a family of four (eighty liters per day total), and a dose of 40 mJ/cm^2 and 75% UVT, a comparison was conducted of current UV-LEDs with current LP systems such as the UV-Tube and the Sterilight (R-Can Environmental Inc., Guelph, Ontario, Canada) systems. The base case includes current UV-LED specifications and assumes a constantly running system. The comparison presented in Table 1 shows the much greater cost of UV-LEDs, both upfront and over time since the lifetime is much lower than the LP systems. However, SET and Crystal IS (Green Island, New York), manufacturers of UV-LEDs, estimate great improvements in the next three to four years. If the projected values manufacturers are aiming for are met, a UV-LED system could be a viable and improved option over current LP systems and fill a gap for low-flow, inexpensive systems in three to four years (Table 1).

Increasing power output will be necessary for systems to utilize a reasonable number of LEDs independent of lamp cost. Each LED requires wiring and other electrical components such as resistors and heat sinking material. More LEDs also require a larger system and more materials that will cost more up front. Maintenance will also be more difficult with a larger number of LEDs since each device will need to be monitored to detect failures. This will be particularly important for systems that require a high flow rate, where thousands of LEDs may become difficult to install and maintain. Based on manufacturer expectations, 100 mW (power output) LEDs should be on the market by 2013. Improving the power output based on manufacturer projections over the next three to four years, shows a large

Table 2 | Effect of improving all three parameters; power output, lifetime, and cost

Vary all	Base case	Case 1	Case 2	Case 3	Case 4	Case 5
MW/lamp	0.36	2	5	10	50	100
Lifetime (hrs)	1,000	2,000	4,000	6,000	8,000	10,000
Cost ($/mW)	664	332	100	10	1	0.1
Total lifetime (days)	42	83	167	250	333	417
Total lifetime (years)	0.11	0.23	0.46	0.68	0.91	1.14
Number of LEDs	201	37	15	8	2	1
Total lamp cost (upfront) USD	47,943	23,975	7,222	722	72	7
Cost for 3 years USD	1,260,063	315,068	47,450	3,163	237	19

decrease in the number of LEDs required (from over 200 to only one LED) for a constantly running household system that would treat eighty liters per day at 40 mJ/cm^2 with a UVT as low as 75%.

One of the most desired features of LEDs for disinfection systems is their long lifetime, particularly for developing communities, where replacements can be difficult to come across. According to SET engineers, UV-LEDs in the germicidal wavelength range currently have very low lifetimes of approximately 1,000 hours before 50% power reduction is reached. Manufacturer projections for the next three to four years would offer lifetimes equal to that of LP lamps (10,000 hours by around 2012). However, since they do not need to warm-up, they can be effectively run intermittently on demand, increasing the total lifetime ten to twenty fold assuming no decay due to frequent on-off switching. LP lamps can be run intermittently as well, but due to the required warm-up time, water is not available on demand without the need for storage, a known hygiene risk.

Because UV LEDs were shown to be as effective for bacterial inactivation as LP UV lamps, the most influential factor to improve the adoption of UV-LED disinfection is cost decrease. Based on manufacturer's three to four year projections, the cost will decrease over 1,000 fold to $0.1 per mW in 2013. The large decrease in three-year cost for a household system brings the total cost to $190 USD, which is almost as cheap as the three year cost for the Sterilight system lamps at $165 USD (Table 1).

Combining projected improvements to power output, lifetime, and cost per mW, results in UV-LEDs being a feasible option and an improvement over LP systems around the year 2013 (Table 2). If the projections can be met, it will be possible to develop a household system that will treat eighty liters per day at 40 mJ/cm^2 (if UVT of water greater than or equal to 75%) for $7 USD of upfront lamp cost, compared to $20 to $55 USD for lamps in the UV-Tube and Sterilight systems, respectively. The cost savings will increase yearly with slightly higher lifetime values of 10,000 hours for the LEDs, versus 9,000 hours for the LP lamps, resulting in lower yearly replacement costs.

The long-term cost savings can be increased further and the maintenance required to replace burned out lights

Table 3 | Effect of increasing flow rate for future UV-LED systems

Vary flow rate	Constant	Case F1	Case F2	Case F3	Case F4	Case F5
Flow rate (mL/min)	55.55	500	1,000	1,890	5,000	6,000
Hours/day	24.0	2.7	1.3	0.7	0.3	0.2
Total lifetime (days)	417	3,750	7,500	14,175	37,500	45,000
Total lifetime (years)	1.14	10.27	20.55	38.84	102.74	123.29
Total mW	72.22	650	1,300	2,457	6,500	7,800
Number of LEDs	1	7	13	25	65	78
Total lamp cost (upfront)	7	65	130	245.7	650	780

can be decreased, by increasing the system flow rate and turning on the LEDs intermittently as water is needed without the need for storage where recontamination can easily occur (Table 3).

CONCLUSIONS

UV-LEDs are an effective technology to inactivate *E. coli* in water, comparable to LP UV mercury vapor lamp technology. The efficiency of UV-LEDs at 265 nm was not found to be statistically different from low-pressure UV sources. A ten LED prototype served as a proof-of-concept for flow-through water treatment, but currently UV-LEDs in the germicidal wavelength range are much too expensive, low power and have short lifetimes. For UV-LEDs to be a feasible option for field implementation the cost needs to decrease and the power output needs to increase substantially. However, according to an analysis of manufacturer projections, UV-LEDs should be a viable and economic option within four years, by around 2013. Once UV-LEDs become a viable option, there are numerous possible applications for UV-LED technology within the water sector, including low-pressure lamp replacement in drinking water, wastewater, and gray water treatment systems, in solar powered or plug-in point-of-use treatment systems in rural and urban households in developing countries, and in portable systems.

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