Electronic Supplementary Material (ESI) for Environmental Science: Water Research & Technology.

Human Health Trade-offs in the Disinfection of Wastewater for Landscape Irrigation: Microplasma Ozonation vs. Chlorination

- Supplementary Information

Shengkun Dong,[1] Jun Li,[1] Min-Hwan Kim,[2, 3] Sung-Jin Park,[2, 3] J. Gary Eden,[2, 3]

Jeremy S. Guest,[1*] and Thanh H. Nguyen[1*]

[1] Department of Civil and Environmental Engineering,

[2] Department of Electrical and Computer Engineering,

University of Illinois at Urbana-Champaign

Urbana, IL 61801, USA

[3] EP Purification, Inc.,

Champaign, IL 61822, USA

[*]Authors to whom correspondence should be addressed:

Thanh H. Nguyen (thn@illinois.edu) and Jeremy S. Guest (jsguest@illinois.edu)

1. Obtaining *L. pneumophila* Inactivation Parameters

The measured dissolved ozone concentrations (C_L) at different times were fitted to the solution to the mass balance equation for the batch reactor:

$$\frac{dC_L}{dt} = -kC_L \qquad S(1)$$

where the fitting parameter is the first order rate constant (k) for ozone decomposition in organic-free solutions. The fitted k values were 5.4 (±0.18) $\times 10^{-3}$ min^{-1} ($R^2 \geq 0.99$) for 22 °C and 1.8 (±0.048) $\times 10^{-3}$ min^{-1} ($R^2 \geq 0.99$) for 7 °C. The results are comparable to those published previously.[1] To determine the volumetric liquid-phase mass transfer coefficient (K_La),[2] ozone was added continuously to the buffered organic-free solution, and C_L was measured at different times. The mass balance for ozone mass transfer and decay in this experiment is given as:

$$\frac{dC_L}{dt} = K_La\left(\frac{C_{gi}}{m} - C_L\right) - kC_L \qquad S(2)$$

where C_{gi} is the ozone concentration at the interphase, and m is the Henry's law constant at different temperatures. The value of K_La was then determined by fitting the measured C_L to the solution to eqn. S(2), to be 3.1 (±1.3) $\times 10^{-3}$ min^{-1} ($R^2 \geq 0.88$) for 22 °C and 9.3 (±5.8) $\times 10^{-3}$ min^{-1} ($R^2 \geq 0.95$) for 7 °C. The decomposition of ozone by WOM (represented by TOC) was thus determined by monitoring the change in C_L in wastewater for a given initial dissolved ozone concentration C_0. The mass balance relation for this set of experiments is:

$$\frac{dC_L}{dt} = -k_2C_Lx \qquad S(3)$$

where x is the reactive portion of the TOC. It is assumed that $C_0 - C_L = x_0 - x$, and that the initial value for x is x_0, where $x_0 = \alpha TOC_0$ [3] and where α is the fraction of organic carbon that exerts ozone demand and TOC_0 is the initial total organic carbon loading. The fraction of

reactive WOM (α) and the reaction rate constant (k_2 in unit of L μg^{-1} min^{-1} were determined by fitting the measured C_L to the solution to eqn. S(3). The fitted parameters are $\alpha = 0.1\ (\pm 0.0)$ mg O_3 (mg C)$^{-1}$ for both 22 ($R^2 \geq 0.72$) and 7 °C ($R^2 \geq 0.58$), and k_2 = 2.9 (± 3.7) ×10^{-2} L μg^{-1} min^{-1} ($R^2 \geq 0.76$) for 22 °C and k_2 = 2.6 (± 1.9) ×10^{-2} L μg^{-1} min^{-1} ($R^2 \geq 0.83$) for 7 °C. All fitting parameters were average values from at least four biological replicates. The values for the second order reaction rate constant are similar to the ones reported in the literature.[4] eqn. S(3) can be solved by substituting x and x_0 into the equation. The overall time rate of change of ozone concentration is thus expressed as:

$$\frac{dC_L}{dt} = K_L a\left(\frac{C_{gi}}{m} - C_L\right) - k_1 C_L y - k_2 C_L(x_0 - C_0 + C_L) - kC_L \qquad S(4)$$

where the term $-k_1 C_L y$ represents the loss of ozone through reacting with bacteria. Experiments examining *L. pneumophila* inactivation in the organic-free solution showed statistically insignificant difference of ozone decay rate with and without the presence of *L. pneumophila* ($p > 0.05$), therefore the term considering bacterial consumption of ozone is negligible in this study, and the final mass balance equation used for parameter fitting is:

$$\frac{dC_L}{dt} = K_L a\left(\frac{C_{gi}}{m} - C_L\right) - k_2 C_L(x_0 - C_0 + C_L) - kC_L \qquad S(5)$$

Terms in the above equation that were measured in the experiments are the dissolved ozone concentration as a function of time (C_L), the initial total organic carbon loading (TOC_0), the initial dissolved ozone concentration (C_0), and the ozone concentration at the interphase (C_{gi}), which is assumed to be equal to the gas phase ozone concentration due to negligible mass transfer resistance in the ozone gas film. A finite difference method was used to solve eqn. S(5) on a time scale as large as 15 minutes, with a time interval of 0.2 or 5 seconds, depending on the time scale of interest. The solution to eqn. S(5) was used to predict the dissolved ozone

concentration under a variety of experimental conditions. The solutions to eqn. S(1) - (3) are provided below:

Solution to eqn. S(1):

$$\mathrm{Ln}\left(\frac{C_L}{C_0}\right) = -kt \qquad \text{S(6)}$$

Solution to eqn. S(2):

$$C_L = \frac{\exp\left(t(K_La+k)\right)K_La\frac{C_{gi}}{m} - K_La\frac{C_{gi}}{m}}{\exp(t(K_La+k))(K_La+k)} \qquad \text{S(7)}$$

Solution to eqn. S(3):

$$C_L = \frac{\exp((-k_2\alpha TOC_0+k_2C_0)t)(-C_0k_2\alpha TOC_0+k_2{C_0}^2)}{\exp\left((-k_2\alpha TOC_0+k_2C_0)t\right)k_2C_0-k_2\alpha TOC_0} \qquad \text{S(8)}$$

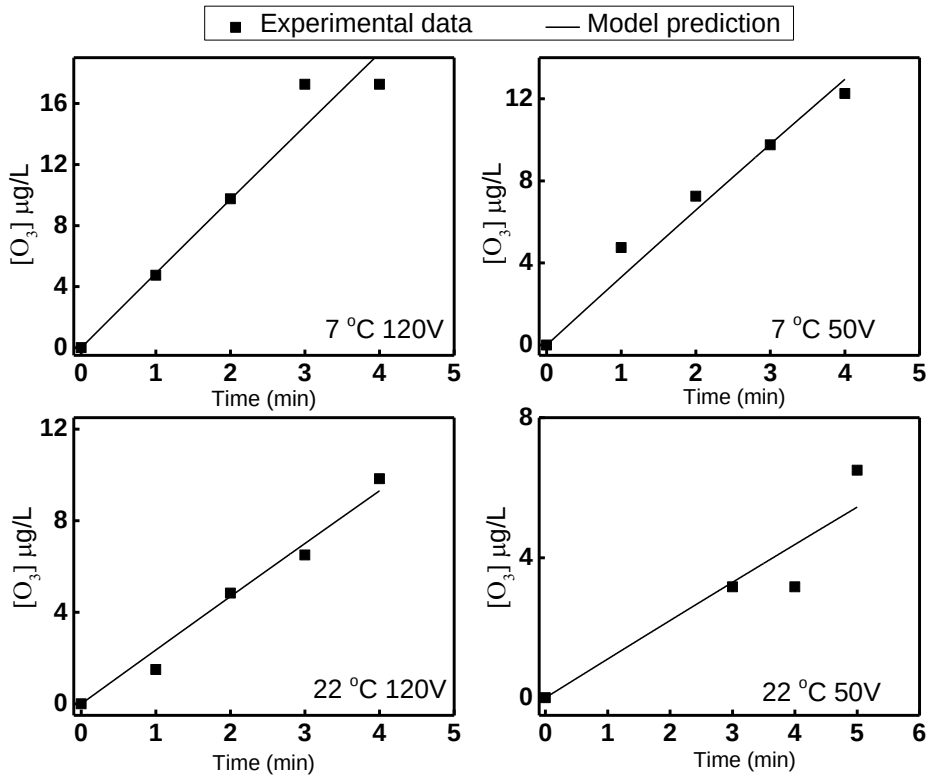

Figure S1. Examples of profile of ozone concentration during constant flow rate purging into buffer solution at pH 6.8 – 7, and at 7 and 22 °C.

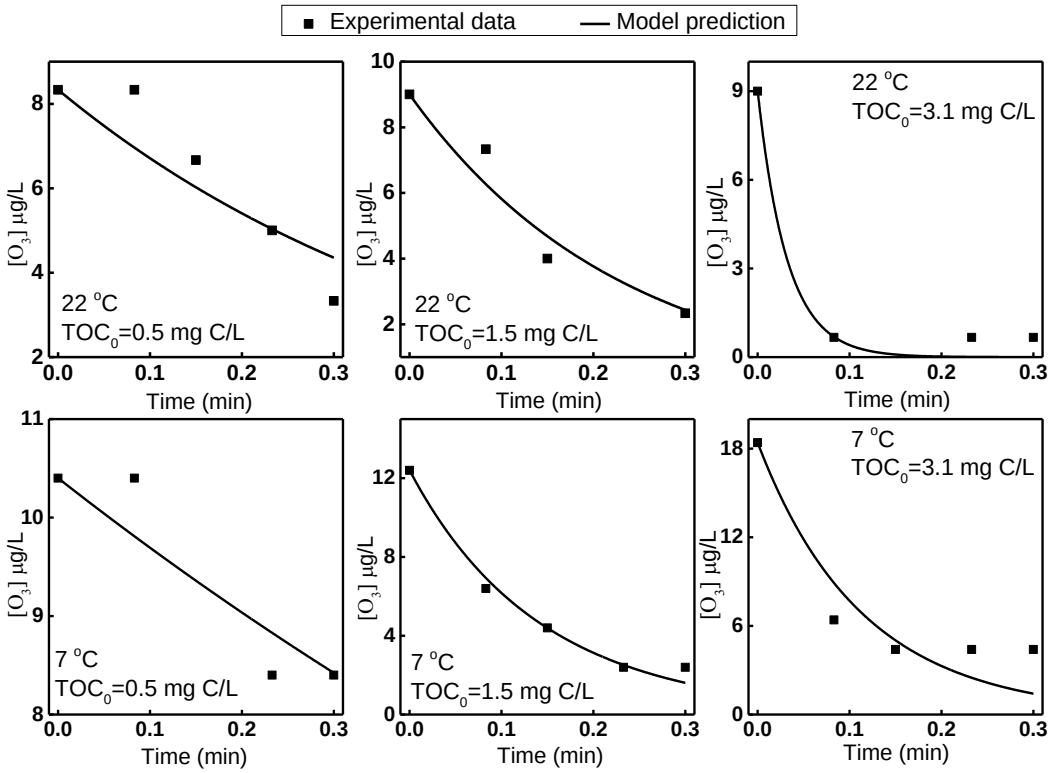

Figure S2. Examples of profile of ozone concentration during ozone reaction with WOM at various initial TOC loadings at pH 6.8 – 7, and at 7 and 22 °C. Due to the complexity of the wastewater matrix and difficulties encountered to precisely capture the ozone decay profiles, uncertainties were involved during the fitting of the reaction rate constant k_2, which was subsequently used in the ozonation system design (see the SI, section 2.3). These uncertainties were accounted for using the Pedigree matrix approach in the uncertainty analysis.

2. Design Process Overview

2.1 General Assumptions

Assumptions were made during the design process when detailed information was not available. The summary of major assumptions is provided in Table S1.

Table S1. Summary of major assumptions for the life cycle assessment.

Assumptions
Fuel sources for electricity in California, Florida, and Texas are based on the US 2015, 2013, and 2013 energy consumption data, respectively
The flow inside the chlorine contact tank follows an ideal plug-flow pattern
The chemical storage tanks and injectors do not need to be replaced in 10 years
Thickness of individual layers of materials of the chemical storage tanks is assumed (later shown to be insensitive to the final results), with the total thickness fixed according to design handbook
Only consumables for treatment operation are included
The flow inside the ozone contact tank follows an ideal plug-flow pattern
Consumption of ozone is entirely incurred by the organics in wastewater
The microplasma ozone generator does not need to be replaced in 10 years
No post ozonation filtration is implemented

2.2 Chlorine Disinfection System Design

The design of chlorination/dechlorination system in general followed the EPA design manual for the municipal wastewater disinfection.[5] The system boundary included chemical storage tanks, pumps, inline static mixers, chlorine contact tank, and consumables (including energy and the production and transport of chemicals to the site). The system was designed to handle 4 Million Gallons per Day (MGD) of secondary effluent, with a HRT of 15 to 45 minutes and a sodium hypochlorite dose of 2 to 8 mg L^{-1}. The designed annual operation temperature range was from 10 to 21.1 ^{o}C with 15.6 ^{o}C as the medium value, which is typical for municipal wastewater.[6]

The decay of sodium hypochlorite was modelled with a parallel decay of two components of total chlorine residual for secondary effluents:[7]

$$C = C_0 e^{-k_1 t} + C_0(1 - x)e^{-k_2 t} \quad \text{S(9)}$$

For a variety of wastewater sources and water conditions, values of $x = 0.3$, $k_1 = 1.67 \times 10^{-2}\ s^{-1}$, and $k_2 = 5 \times 10^{-5}\ s^{-1}$ yielded satisfactory results.[5] Since a hydraulic residence time (HRT) of 15 to 45 minutes is typical for wastewater chlorination disinfection, which is also the design HRT range for the current study, for HRT > 1 minute, eqn. S(9) can thus be re-written as:[5]

$$C = 0.7C_0 e^{-0.003t} \quad \text{S(10)}$$

Combine eqn. S(5) with Chick-Watson inactivation model (eqn. S(16)) and integrate, we arrive at:

$$\frac{N}{N_0} = e^{[-0.7kC_0(333 - 333e^{-0.003t})]} \quad \text{S(11)}$$

where k is the inactivation rate constant for pathogens, and C_0 is the applied chlorine dose. The inactivation rate constants for *Giardia*, *Cryptosporidium parvum*, and *Legionella pneumophila* are 2.5×10^{-2} (at 18 ^oC), 8.36×10^{-4} (at 20 ^oC), and 0.307 (at 30 ^oC) L mg^{-1} min^{-1}, respectively.[8-10] The temperature effect was considered by incorporating the Arrhenius correction:

$$k_{T_2} = k_{T_1} e^{\frac{E_a}{R}\left(\frac{1}{T_1} - \frac{1}{T_2}\right)} \quad \text{S(12)}$$

where E_a is the activation energy in J mol^{-1}, R is 8.314 J mol^{-1} K^{-1}. T_1 and T_2 are temperatures before and after temperature correction. Whenever not directly available, E_a values were fitted

from published experimental data. The E_a values for *Giardia*, *C. parvum*, and *L. pneumophila* used in this study are 45259, 75570, and 38293 J mol^{-1}, respectively. The activation energy for *L. pneumophila* was estimated based on monochloramine inactivation data of eight species of bacteria.[11]

The chlorine contactor was designed following a length to width ratio of 40 to 1, to minimize axial diffusion.[5, 6] The depth of the contactor was empirically chosen based on the EPA design manual as 2 meters, with an effective depth of 1.8 meters. The horizontal velocity of wastewater inside the contact tank ranged from 2 – 4 m min^{-1}.[6] Since the influent flow rate was known, given the horizontal velocity the cross sectional area of the tank and thus width of the tank were calculated. Length was calculated given the width and the length to width ratio. The final volume of materials was determined by applying the density of the concrete.

The storage, injection, and mixing of the sodium hypochlorite and sodium bisulfite shared similar configuration as the equipment is usually interchangeable. Exceptions were the materials for the storage tanks and the wetted parts of the feed pumps, as sodium hypochlorite is highly corrosive. Information on the storage tanks were obtained from the design handbook.[12] The masses of components and materials of the proportioning feed pumps were obtained from manufacture's specification sheets, which were scaled up or down according to the pump's horsepower. Mixing of both the sodium hypochlorite and sodium bisulfite were done with static inline mixers. Since the dechlorination reactions happen instantaneously, contact chambers for dechlorination was not required.[6]

The usage of sodium hypochlorite and sodium bisulfite was calculated based on the reactions:

$$NH_2Cl + HSO_3^- + H_2O \rightarrow Cl^- + SO_4^{2-} + NH_4^+ + H^+ \qquad \text{REACTION S1}$$

$$HOCl + HSO_3^- \rightarrow Cl^- + SO_4^{2-} + 2H^+$$ REACTION S2

The final results calculated from stoichiometry was further combined with 12% empirical safety factors to yield a ratio of 1.63 mg L^{-1} $NaHSO_3$ per 1 mg L^{-1} residual chlorine as Cl_2.[6]

The requirements for the sodium hypochlorite and sodium bisulfite were calculated following the design manual.[5] In brief, the mass rate requirements were calculated using the stoichiometry, the influent flow rate, and the applied chemical dose. The volume of chemicals to be stored onsite was calculated following the EPA recommendation that utilities should have the volume of chemicals that equal the amount of chemicals needed for a time equal to the shipping time from the vendor plus fifteen days. A 4-day shipping time was assumed as the base value for the uncertainty analysis.

2.3 Microchannel Plasma Ozonation Disinfection System Design

The design of microplasma ozonation system was based on a combination of both scaled-up results from bench-scale studies, manufacture's data and design criteria from the design manuals and previous publications. The system boundary included the ozone generator and injection device, ozone contact tank, and the ozone destruction device. Identical with the chlorination system, the microplasma disinfection system was designed to handle 4 MGD of secondary effluent, with a HRT of 7 to 11 minutes and a transferred ozone dose of 1 to 2 mg L^{-1}. The operational temperature range is identical with the chlorination system, from 10 to 21.1 ^{o}C.

The decay of ozone caused by various components of the secondary effluent was addressed as follows. First of all, it was assumed that Total Organic Carbon (TOC) is a good representation of the components of wastewater that could exert ozone demand.[3] Since the representative TOC concentration was not readily available for secondary effluent, a correlation between TOC and Chemical Oxygen Demand (COD) was used following a prior publication:

$$\mathrm{COD} = 7.25 + 2.99\mathrm{TOC} \qquad \text{S(13)}$$

which was developed for the final effluent.[13] The designed COD concentration for the microplasma ozonation unit's receiving stream was 30 to 35 mg L^{-1}, which is typical for secondary effluent.[6] The decomposition of ozone, which includes its self-decomposition and the reaction with organic matter in wastewater (represented by TOC) was thus modeled for a batch system as:

$$\frac{dC_L}{dt} = -k_2 C_L x - kC_L \qquad \text{S(14)}$$

where C_L is the transferred ozone dose in mg/L, and x is the reactive portion of the TOC. It is assumed that $C_0 - C_L = x_0 - x$, and that the initial value for x is x_0, where $x_0 = \alpha TOC_0$. Also, α is the fraction of organic carbon that exerts ozone demand. k_2 is the second order reaction rate constant between ozone and TOC, and k is the first order reaction rate constant for ozone self-decomposition. The values for k_2 and k were obtained from the bench-scale experiments conducted in this study. To correct for temperature variations, eqn. S(12) was applied and the activation energy was 5005 and 50350 J mol^{-1} for k_2 and k, respectively. After integration, the solution to eqn. S(14) is:

$$C_L = \frac{x_0 k_2 C_0 - k_2 {C_0}^2 + C_0 k}{(x_0 k_2 + k) e^{t(x_0 k_2 - k_2 C_0 + k)} - k_2 C_0} \quad \text{S(15)}$$

Combine eqn. S7 with Chick-Watson equation (eqn. S(16)), integrate, one can arrive at the equation to predict level of inactivation with ozone produced from the microplasma ozone generator (eqn. S(17)):

$$\frac{dN}{dt} = -k^* C_L N \quad \text{S(16)}$$

$$\text{Ln}\frac{N}{N_0} = \frac{k^*}{k_2}\left\{t[k_2(x_0 - C_0) + k] - \text{Ln}\frac{(k_2 x_0 + k) e^{t(k_2 x_0 - k_2 C_0 + k)} - k_2 C_0}{k_2 x_0 + k - k_2 C_0}\right\} \quad \text{S(17)}$$

where N and N_0 are the concentration of pathogens in wastewater at time t and at time 0, respectively. The ozone inactivation rate constants for *Giardia*, *C. parvum*, and *L. pneumophila* are 27.1 (at 25 ^oC), 0.8 (at 20 ^oC), and 145 (across 7 to 22 ^oC) L mg^{-1} min^{-1}, respectively.[8, 14] To correct the rate constants for temperature variations, the Arrhenius rule (eqn. S(12)) was applied, where the activation energy E_a was 39201 and 82500 J mol^{-1} for *Giardia* and *C. parvum*, respectively.[8, 14] The only exception for the temperature correction for rate constants is the inactivation rate constant for *L. pneumophila*, which was demonstrated to be not significantly

sensitive to temperature variation in the presence of wastewater organic matter, based on the good fitting to the Chick-Watson equation at different temperatures (Figure. 1).

Ozone generated from the microplasma ozonator was designed to be injected into the wastewater through a venturi injector. To obtain the number of the microplasma chips (n), which are the very components that control the total ozone mass production rate of the microplasma ozonator, as well as the factors that directly determine the material requirement of the ozone generator, eqn. S(18) was applied, which was derived based on the venturi injector manufacture's design document on the relationship between MTE and ozone demand for the 87% MTE venturi injector that the current study uses (eqn. S(19)). Ozone demand was empirically predicted using an equation that was fitted from data provided in the EPA design manual (eqn. S(20)):

$$n = (100C_0 - 13 \times \text{Ozone demand}) \frac{Q_{Lin}}{87 \times \dot{O}_{3|\text{per piece}}} \quad \text{S(18)}$$

$$\text{MTE}(\%) = (13 \frac{\text{ozone demand}}{\text{applied ozone dose}} + 87) \quad \text{S(19)}$$

$$\text{Ozone demand} = 0.0844 \times \text{COD} - 1.3698 \quad \text{S(20)}$$

where the applied ozone dose is defined in eqn. S(21):

$$\text{Applied ozone dose} = \frac{Q_{gin} C_{gin}}{Q_{Lin}} \quad \text{S(21)}$$

The ozone contact tank shared a similar configuration with the chlorine contact tank,[12] with the addition of a concrete cover to collect and pass the off-gas to the thermal catalytic ozone destruction unit. The thermal catalytic ozone destruction unit contains MnO_2 and CuO as catalysts and operates at 300 to 350 ^oC to remove approximately 99% ozone.

3. Calculating DALYs per Symptomatic Case for *L. pneumophila*

Legionellosis is a common syndrome of pneumonia caused by *Legionella*,[15] and the bacterium *L. pneumophila* is responsible for most cases of Legionellosis.[16] 1- 40% of hospital acquired pneumonia can be attributed to Legionellosis,[17] and 95.4 % of Legionellosis is caused by *L. pneumophila*.[18] Therefore, the same weighting was assumed to be reflective of the proportion of pneumonia DALYs attributable to Legionellosis.

Based on RIVM, The Netherland (Table S2.), the total amount of people affected by pneumonia nationwide in a year was 645584. Therefore DALYs per symptomatic case can be determined as:

$$\frac{73900}{6984+638600} = 0.1145 \text{ DALYs per symptomatic case} \qquad S(22)$$

As discussed 1- 40% of hospital acquired pneumonia is attributed to Legionellosis, and 95.4 % of Legionellosis is caused by *L. pneumophila*, it can therefore be assumed that the DALYs/symptomatic case for *L. pneumophila* infection is within the range of 1.05×10^{-3} to 4.37×10^{-2}. This source of uncertainty was incorporated into the uncertainty analysis.

Table S2. Yearly population affected by pneumonia in The Netherland.

	# Deaths	**Years of life lost (YLL)**	**# Disease**	**Severity**	**Years of Life with Disability (YLD)**	**DALYs**
Pneumonia	6984	49448	638600	0.04	24500	73900

Table S3. Electricity consumption source profiles in percentage (%) for three states within the United States with the highest reclaimed water usage. Distillate fuel oil was represented with light fuel oil. Motor gasoline was represented with unleaded petrol. Other petroleum was represented with 15% (v/v) ETBE added petroleum. Other renewable energy sources were represented with biofuel.

Electricity fuel sources based on consumption	**Ecoinvent 3 database items**	**Florida**	**California**	**Texas**
Natural gas	Electricity, natural gas, at power plant/US	31	61	32
Nuclear	Electricity, nuclear, at power plant/US	7	10	3
Hydroelectric	Electricity from hydroelectric power plant, AC, production mix, at power plant, < 1kV RER S	0	5	0
Coal	Electricity, bituminous coal, at power plant/US	12	0	12
Liquefied petroleum gas	Liquefied petroleum gas {RoW}| market for | Alloc Def, U	0	0	15
Distillate fuel oil	Light fuel oil {RoW}| market for | Alloc Def, U	7	0	7
Motor gasoline	Petrol, unleaded {RoW}| market for | Alloc Def, U	23	0	11
Other Petroleum	Petrol, 15% ETBE additive by volume, with ethanol from biomass {GLO}| market for | Alloc Def, U	1	0	10
Other renewable	Electricity, biomass, at power plant/US	8	22	0

Table S4. Summary of input materials for the chlorination system.

Summary of input materials for chlorination system															
Materials	**Processes**	Storage tanks		Piping assembly				Contact tank	Pump		Consumables (annual requirements)		Electricity (annual requirements)		
		For NaOCl storage (kg)	For $NaHSO_3$ storage (kg)	Injection pipe for NaOCl (kg)	Injection pipe for $NaHSO_3$ (kg)	Inline static mixing pipe for NaOCl (kg)	Inline static mixing pipe for $NaHSO_3$ (kg)	Contact tank (m^3)	For NaOCl (kg)	For $NaHSO_3$ (kg)	NaOCl (kg)	$NaHSO_3$ (kg)	NaOCl feed pump (kWh)	$NaHSO_3$ feed pump (kWh)	Coefficient of variation
Fiberglass		1353.6													0.23
PVC lining		1052.8													0.23
316 SS			6044.5		46.1	171.8	171.8		27.6	27.6					0.10
PTFE									0.3	0.3					0.11
Copper wire									0.6	0.6					0.23
Steel									5.8	5.8					0.23
Schedule 80 PVC				8.1											0.36
Slab concrete								42.0							0.23
$NaHSO_3$ powder												22981.6			0.03
15% NaOCl solution											177719.4				0.03
Electricity, low voltage													952.9	411.5	0.04
	Blow moulding	1052.8													0.36

Table S5. Summary of input materials for the microplasma ozonation system.

Summary of input materials for microplasma ozonation system									
	Ozone generator	Piping assembly			Contact tank	Ozone destruction unit	Electricity		
Materials	Ozone generator (kg)	Injection pipe (kg)	Venturi (kg)	Equalization pipe (kg)	Contact tank (m^3)	Ozone destruction unit (kg)	Ozone generator electricity (kWh)	Ozone destruction unit electricity (kWh)	Coefficient of variation
316 SS	14.7	46.1	151.1	414.9		0.05			0.10
Copper wire	9.8								0.23
Slab concrete					23.6				0.23
Aluminum Foil	163.3								0.01
Titanium dioxide	1.0								0.21
Silicon	0.1								0.01
Polycarbonate	22.5								0.01
PVC for Fan Blades	2.9								0.03
Printed Circuit board-Pb containing	0.1								0.01
Printed Circuit board-No Pb	0.1								0.01
MnO_2						0.01			0.04
CuO						0.01			0.04
Electricity, low voltage							94224.8	2.2	0.04

Table S6. Input parameters for the life cycle analysis and quantitative microbial risk assessments.

								Distribution		
Parameter	Unit	Minimum	Maximum	Mean for log normal distribution	Standard deviation for log normal distribution	Mean	Standard deviation	Uniform	Log normal	Normal
Temperature	^{o}C			-	-	15.6	1.4			•
Influent COD	mg L^{-1}	30	35	-	-	-	-	•		
Cryptosporidium parvum concentration in influent	oocysts mL^{-1}	0.000013	0.00013	-	-	-	-	•		
Legionella pneumophila concentration in influent	cells mL^{-1}	900	1000	-	-	-	-	•		
Giardia concentration in influent	cysts mL^{-1}	0.000007	0.00018	-	-	-	-	•		
Initial chlorine dose	mg L^{-1} as Cl_2	2	8	-	-	-	-	•		
Hydraulic residence time in chlorine contact tank	min	15	45	-	-	-	-	•		
Horizontal velocity in chlorine contact tank	m min^{-1}	2	4	-	-	-	-	•		
Transferred ozone dose	mg L^{-1}	1	2	-	-	-	-	•		
Hydraulic residence time in ozone contact tank	min	7	11	-	-	-	-	•		
Horizontal velocity in ozone contact tank	m min^{-1}	2	4	-	-	-	-	•		
Identical survival probability for *Cryptosporidium parvum*	-	-	-	-5.475054545	0.362099799	-	-		•	
Identical survival probability for *Legionella pneumophila*	-	-	-	-2.815078774	0.399245315	-	-		•	
Identical survival probability for *Giardia*	-	-	-	-3.917035547	0.256506595	-	-		•	
DALYs/symptomatic case for *Legionella pneumophila*	DALYs per symptomatic case	0.00105	0.0437	-	-	-	-	•		
Chlorine inactivation rate constant for *Cryptosporidium parvum*	L mg^{-1} min^{-1}	-	-	-7.086881945	0.406	-	-		•	
Chlorine inactivation rate constant for *Legionella pneumophila*	L mg^{-1} min^{-1}	-	-	-1.180907531	0.406	-	-		•	
Chlorine inactivation rate constant for *Giardia*	L mg^{-1} min^{-1}	-	-	-3.688879454	0.406	-	-		•	
NaOCl shipping time	day	-	-	1.386294361	0.458257569	-	-		•	
Thickness of chlorine storage tank and contactor	m	-	-	-2.574393816	0.458257569	-	-		•	
$NaHSO_3$ shipping time	day	-	-	1.386294361	0.458257569	-	-		•	
Specific energy consumption for chlorine feeding pump	kWh m^{-3}	-	-	1.93441577	0.037416574	-	-		•	

Table S6. (cont.)

Specific energy consumption for $NaHSO_3$ feeding pump	kWh m^{-3}	-	-	1.93441577	0.037416574	-	-		•	
Ozone self-decay rate constant	min^{-1}	-	-	-5.221356325	0.352987252	-	-		•	
Ozone reaction rate constant with organic matter	L mg^{-1} min^{-1}	-	-	3.36729583	0.352987252	-	-		•	
Ozone inactivation rate constant for *Cryptosporidium parvum*	L mg^{-1} min^{-1}	-	-	-0.223143551	0.405709256	-	-		•	
Ozone inactivation rate constant for *Legionella pneumophila*	L mg^{-1} min^{-1}	-	-	4.976733742	0.352987252	-	-		•	
Ozone inactivation rate constant for *Giardia*	L mg^{-1} min^{-1}	-	-	3.299533728	0.405709256	-	-		•	
Fraction of ozone demand per carbon of organic matter	-	-	-	-1.560647748	0.352987252	-	-		•	
Ozone destruction unit specific energy consumption	Wh m^{-3}	-	-	1.549687908	0.043588989	-	-		•	
Pipe thickness (percent of diameter)	-	-	-	-2.525728644	0.458257569	-	-		•	
Cryptosporidium parvum and *Giardia* ingestion amount per exposure	mL	-	-	0	0.096953597	-	-		•	
Exposure frequency for all three pathogens	events $year^{-1}$	-	-	3.951243719	0.096953597	-	-		•	
Exposure duration per event for *Legionella pneumophila* calculation	hr	-	-	-0.693147181	0.458257569	-	-		•	
Wastewater generated per person per day	Gallons $person^{-1}$ day^{-1}	-	-	4.605170186	0.127279221	-	-		•	
Inhalation rate	m^3 hr^{-1}	-	-	-0.328504067	0.091241438	-	-		•	
Retention fraction of *Legionella pneumophila* in lungs	-	-	-	-0.693147181	0.21937411	-	-		•	
Consumables' transportation distance	km	-	-	3.912023005	0.458257569	-	-		•	

Table S7. Urbana Northeast Wastewater Treatment Plant secondary effluent characteristics.

Water quality parameter	Unit	Value
pH	-	7
TOC	mg C/L	8
SUVA	$L\ mg^{-1}\ m^{-1}$	1.8
UV_{254}	m^{-1}	14.4
COD	mg/L	20
Ammonia nitrogen	mg N/L	5.5

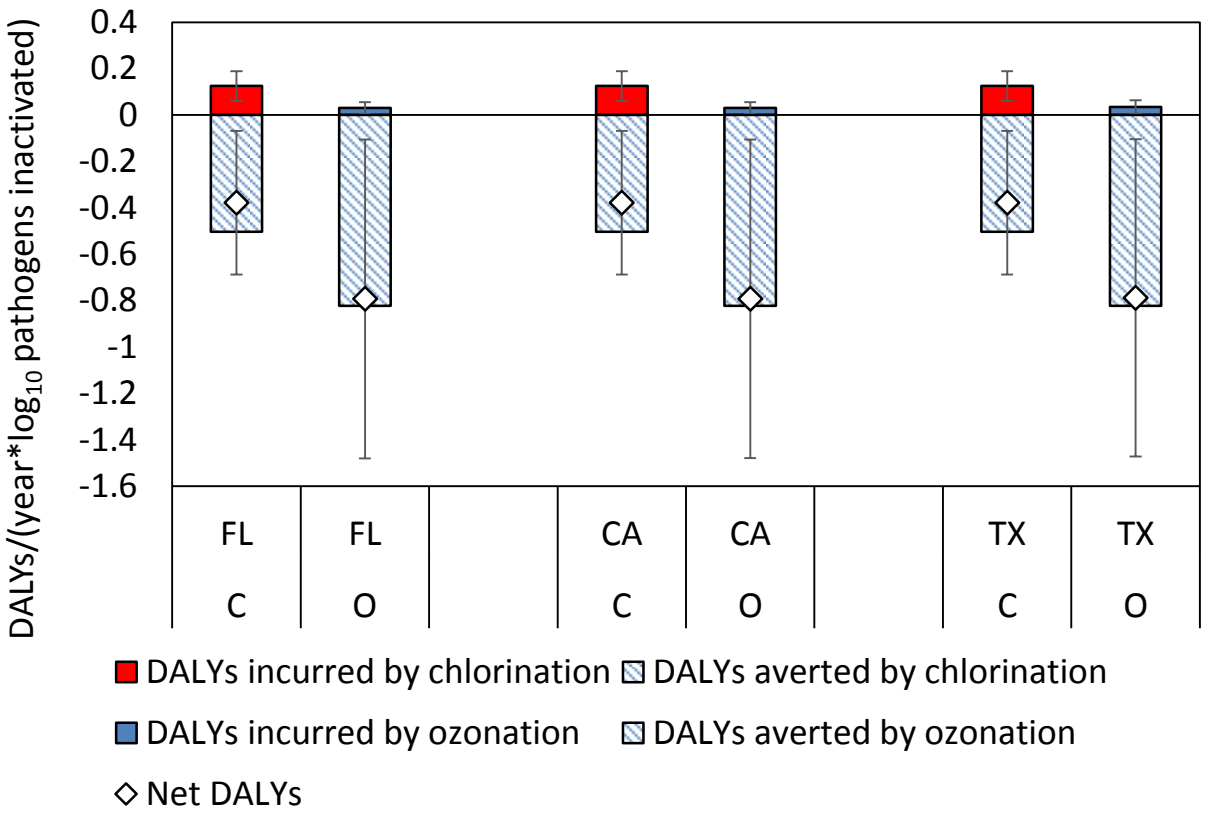

Figure S3. The impact of energy source fuel mix for consumed electricity, expressed in DALYs caused and averted by the disinfection systems, together with the total net DALYs , for the states of Florida, California, and Texas. Abbreviations: FL = Florida, CA = California, TX = Texas, C = Chlorination (combined with dechlorination using sodium bisulfite), O = Microplasma ozonation.

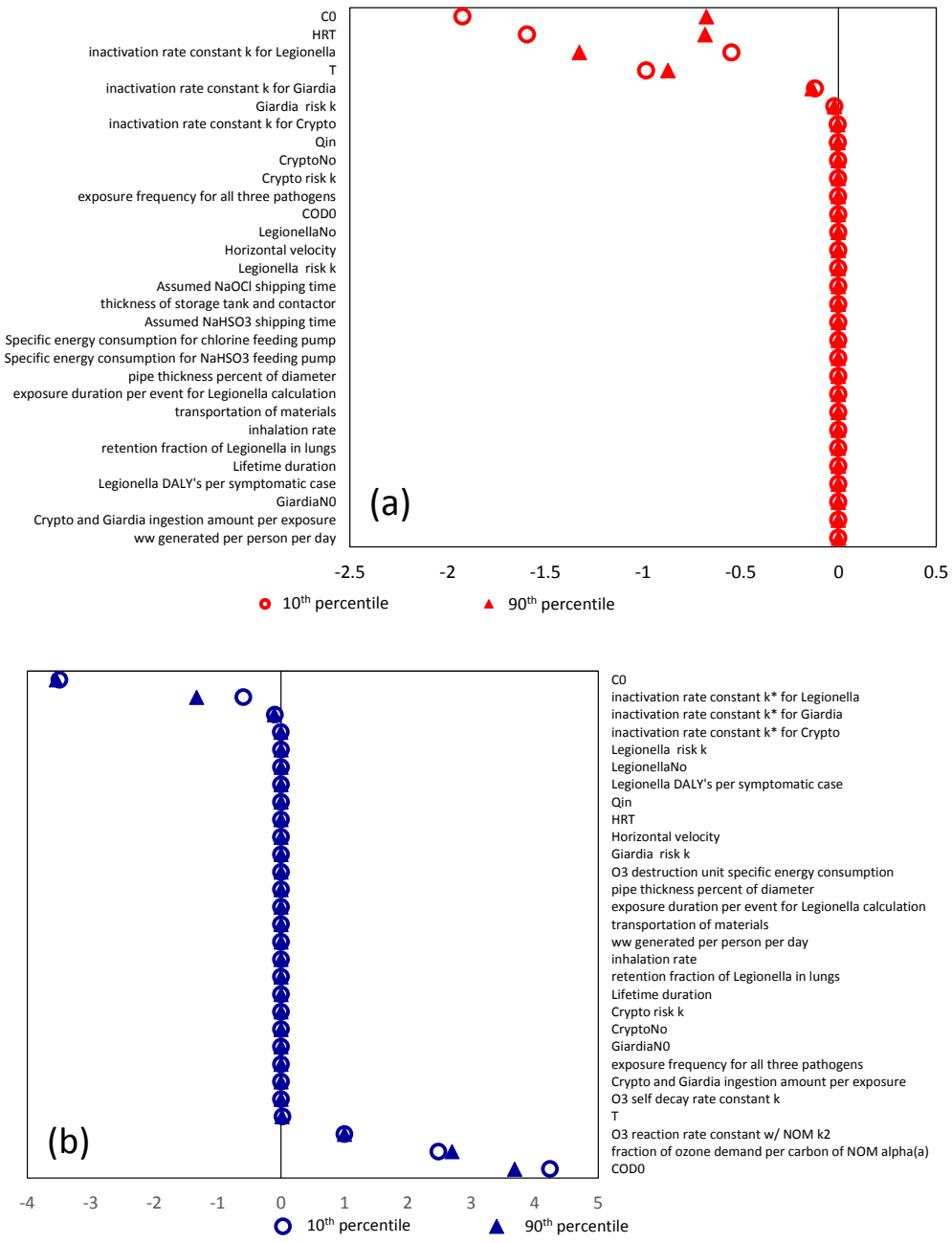

Figure S4. Sensitivity of human health impact to input parameters for chlorination (a) and microplasma ozonation (b) expressed in relative response. Negative values indicate direct correlation between the input values and the output values, meaning that an increase in these input parameters will result in an increase in the overall normalized net DALYs values (less human health protection).

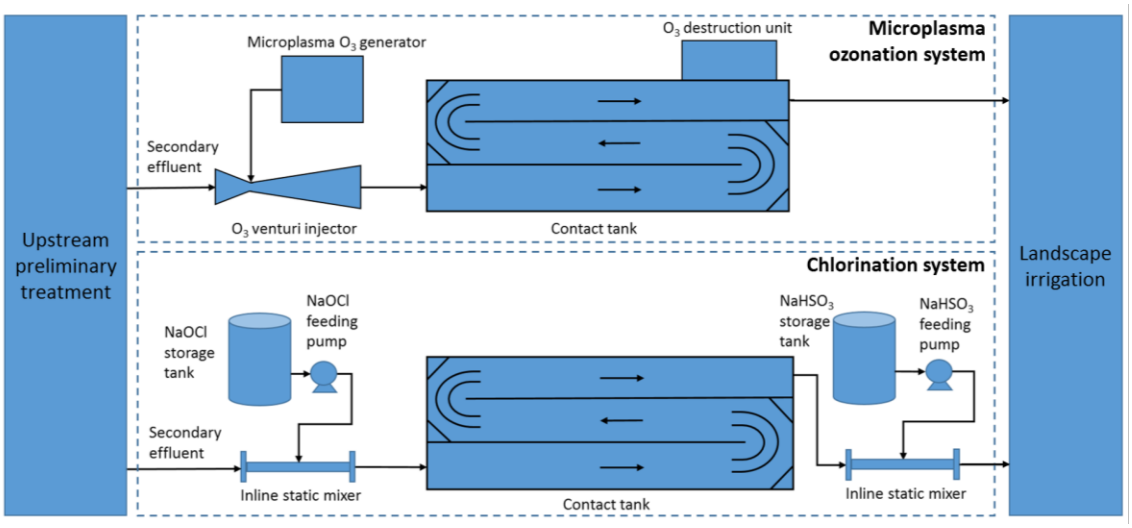

Figure S5. Disinfection system components and LCA system boundaries (dashed lines) of the (top) chlorination system and (bottom) microplasma ozonation system. The construction and operation of the systems were included in the analysis.

References

1. Kilpatrick, M. L.; Herrick, C. C.; Kilpatrick, M., The Decomposition of Ozone in Aqueous Solution1, 2. *J. Am. Chem. Soc.* **1956,** *78*, (9), 1784-1789.
2. Akita, K.; Yoshida, F., Gas holdup and volumetric mass transfer coefficient in bubble columns. Effects of liquid properties. *Ind. Eng. Chem. Proc. Design Devel.* **1973,** *12*, (1), 76-80.
3. Hunt, N. K.; Mariñas, B. J., Inactivation of *Escherichia coli* with ozone: chemical and inactivation kinetics. *Water Res.* **1999,** *33*, (11), 2633-2641.
4. Xu, P.; Janex, M.-L.; Savoye, P.; Cockx, A.; Lazarova, V., Wastewater disinfection by ozone: main parameters for process design. *Water Res.* **2002,** *36*, (4), 1043-1055.
5. Stover, E. L., *Design manual: municipal wastewater disinfection*. U.S. Environmental Protection Agency, Center for Environmental Research Information: 1986.
6. Tchobanoglous, G.; Burton, F.; Stensel, D., *Wastewater Engineering (Treatment, Disposal and Reuse)*. Metcalf and Eddy: New York, 1991; Vol. 1334.
7. Haas, C. N.; Karra, S., Kinetics of wastewater chlorine demand exertion. *J. Water Pollut. Control Fed.* **1984**, 170-173.
8. Jarroll, E. L.; Hoff, J. C., Effect of disinfectants on Giardia cysts. *Crit. Rev. Env. Sci. Tec.* **1988,** *18*, (1), 1-28.
9. Driedger, A. M.; Rennecker, J. L.; Mariñas, B. J., Inactivation of Cryptosporidium parvum oocysts with ozone and monochloramine at low temperature. *Water Res.* **2001,** *35*, (1), 41-48.
10. Cunliffe, D., Inactivation of Legionella pneumophila by monochloramine. *J. Appl. Bacteriol.* **1990,** *68*, (5), 453-459.
11. Rose, L. J.; Rice, E. W.; Hodges, L.; Peterson, A.; Arduino, M. J., Monochloramine inactivation of bacterial select agents. *Appl. Environ. Microbiol.* **2007,** *73*, (10), 3437-3439.
12. Crittenden, J. C.; Trussell, R. R.; Hand, D. W.; Howe, K. J.; Tchobanoglous, G., *MWH's Water Treatment: Principles and Design*. John Wiley & Sons: 2012.
13. Dubber, D.; Gray, N. F., Replacement of chemical oxygen demand (COD) with total organic carbon (TOC) for monitoring wastewater treatment performance to minimize disposal of toxic analytical waste. *J. Environ. Sci. Health A* **2010,** *45*, (12), 1595-1600.
14. Rennecker, J. L.; Mariñas, B. J.; Owens, J. H.; Rice, E. W., Inactivation of Cryptosporidium parvum oocysts with ozone. *Water Res.* **1999,** *33*, (11), 2481-2488.
15. Hilbi, H.; Jarraud, S.; Hartland, E.; Buchrieser, C., Update on Legionnaires' disease: pathogenesis, epidemiology, detection and control. *Mol. Microbiol.* **2010,** *76*, (1), 1-11.
16. Centers for Disease Control and Prevention, Etiologic Agent and Transmission of Legionella. In 2015.
17. Diederen, B., Legionella spp. and Legionnaires' disease. *J. Infect.* **2008,** *56*, (1), 1-12.
18. Marston, B. J.; Lipman, H. B.; Breiman, R. F., Surveillance for Legionnaires' disease: risk factors for morbidity and mortality. *Arch. Intern. Med.* **1994,** *154*, (21), 2417-2422.