Supplementary Material for:

Evaluation of Alternative-to-Gas Chlorination Disinfection Technologies in the Treatment of Maltese Potable Water

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NB. Reference numbering is in accordance with that of the Main Text.

Text S1 Chlorine treatment of the Maltese tap-water

The Maltese potable water is subjected to traditional chlorination for disinfection prior to reticulation and/or distribution. Chlorine gas (supplied in cylinders) is fed to a chlorinator (direct mounting) which comprises a pressure regulating valve, and a regulator that is preceded with a rotameter, for chlorine gas flow regulation. Chlorine flows to the incoming water through vacuum generated by the injector (Figure S1).

Text S2. Preparation of bacterial suspensions

Loopfuls of frozen glycerol stocks were streaked onto tryptic soy agar plates (TSA; Scharlab, Spain) and streaked plates were incubated at 37°C for 16-18 hrs (*E. coli*) or for 36-48 hrs (*E. faecalis*). Confirmation of the species identity was performed by subculturing *E. coli* in chromogenic *E. coli*/coliform agar (Condalab, Spain) followed by completion of oxidase assays using 1% (v/v) tetramethyl-p-phenylenediamine dihydrochloride [34] (Alfa Aesar, UK),

and *E. faecalis* in bile esculin azide agar (Carl Roth GmbH, Germany). After the verification of the species identity, single colonies were used to inoculate TSB media (100-300 mL in 1 L Duran bottles or Erlenmeyer flasks for adequate aeration) which were then incubated at 37ºC overnight (*E. coli*), or for 36-48 hours (*E. faecalis*). After incubation, cells were harvested at 3500 rpm, 20ºC, for 20 min (Rotina 380R; Hettich, Germany). Collected pellets were washed with Ringer's solution (Oxoid, UK) and centrifuged under the same conditions. This washing step was repeated twice. Finally, collected pellets were resuspended in Ringer's solution (100-300 mL) and 1% (v/v) of the suspension was used to spike the desired volume of autoclaved/filtered deionised/RO water prior to microbial inactivation studies.

Text S3. UVC fluence and minimum exposure time calculations

For assessing the bactericidal efficiency of the Sita UVC lamp, Log_{10} inactivations should be examined per applied UVC dose. UVC dose, or *Fluence*, is the time integral of the fluence rate. Fluence is defined as the total radiant energy of a specific light quality and quantity reaching all directions, as the light passes through an infinitesimally small cross sectional area. Assuming a constant and radial distribution of the incident light power (*Irradiance*) and since the lamp output has a clear maximum in the germicidal range (almost resembling the qualities of completely monochromatic light), the UVC Fluence can be calculated from Equation S1.

$$UVC\ Fluence = \frac{Irradiance\left(\frac{W}{m^2}\right)}{Transmittance} \times Exposure\ time \qquad \text{(S1)}$$

Where: m^2 corresponds to the cross-sectional area of the tube. A tube, with diameter *D* (0.05 m), and a distance from the surface of the slieve *t* (0.015 m) (Figure S1) would posses a cross-sectional area that can be calculated using Equation S2.

$$Cross-sectional\ Area = 0.25 \times \pi \times (D^2 - (D - 2 \times t)^2) = \qquad \text{(S2)}$$
$$= 0.25 \text{ x } 3.14 \times (0.050^2\ m^2 - (0.050\ m - 2 \text{ x } 0.015\ m)^2) =$$
$$= 0.25 \text{ x } 3.14 \times (0.0025\ m^2 - (0.050\ m - 0.030\ m)^2) =$$
$$= 0.25 \text{ x } 3.14 \times (0.0025\ m^2 - 0.0004\ m^2) =$$
$$= 0.25 \text{ x } 3.14 \times 0.001204\ m^2 =$$
$$= 0.00165\ m^2$$

$$Power = 3.311\ \text{V} \times 0.02\ \text{A} = 0.06622\ \text{W}$$

$$Irradiance = \frac{0.06622\ \text{W}}{0.00165\ \text{m}^2} = 39.9\frac{\text{W}}{\text{m}^2} = 39.9\frac{\text{J}}{\text{s m}^2}$$

For establishing minimum exposure times, it is firstly necessary to subtract the volume of the slieve surrounding the UVC lamp from the total volume of the UVC chamber. The difference will unveil the volume which is available and can be occupied by the water entering the device. For example, in the case of the set-up (Figure S2):

$$Volume_{chamber} - Volume_{slieve} = 0.00129\ \text{m}^3 = 1.29\ \text{L}$$

Then, to determine the minimum exposure time at a specific flow rate Equations S3 and S4 were used [32].

$$\text{Turbulent flow: } t_{RMin} = 0.82\ t_{AV} \quad \text{(S3)}$$

$$\text{Laminar Flow: } t_{RMin} = 0.50\ t_{AV} \quad \text{(S4)}$$

Where: t_{AV} is the average velocity (flow) and t_{RMin} is the minimum exposure time. Then, at a flow of e.g., 0.6 L/min, the t_{RMin} for the laminar flow would be:

$$t_{RMin} = 0.50\ t_{AV} = 0.50 \times 0.60\ \text{L/min} = 0.3\ \text{L/min}$$

However, since the capacity of the chamber is 1.29 L, the actual t_{RMin} for the laminar flow, woulld be:

$$t_{RMin} = 1.29\ L\ \times \frac{1\ min}{0.3\ L} = 4.3\ \text{min} = 258\ s$$

Thus, for this minimum exposure time, the UVC fluence would be:

$$UVC\ Fluence = \frac{Irradiance}{Transmittance} \times Exposure\ time =$$

$$= \frac{39.9\ \left(\frac{J}{s\ m^2}\right)}{100\%} \times 258\ s = 10.1\ \frac{kJ}{m^2}$$

With maximum transmittance, maximum intensity is achieved. Changes in transmittance, would be reflected in the recorded percentage intensity by the control panel. In that case, the UVC fluence should be corrected by multiplying with the appropriate factor. For example 73% maximum Intensity, should affect the voltage by 0.73x, resulting in an actual fluence of 7.3 kJ/m^2. With UVC fluences calculated (Table S1), Log_{10} bacterial inactivations were plotted against the applied fluences, and the inactivation models of GinaFit were used to fit the data (Text S4).

Text S4. GinaFit kinetic models used in the current study

The bacterial inactivation profiles for the applied treatments exhibited biphasic characteristics, with a fast inactivation phase showing dependence on the applied fluence and/or dose, and a slower phase often characterised by the presence of a tail, indicative of the presence of a residual bacterial population surviving the treatment. None of the observed bacterial inactivation profiles exhibited "shoulder-like" periods before the exponential reduction. The models which best described the obtained data per applied technology are described.

Text S.4.1 UVC; The modified Weibull equation [36] accounting for tailing effects stemming from the presence of putative resistant-to-the treatment populations and adapted to reflect fluence/dose dependencies (Izguier and Gómez-López, 2011, cited in [37]) was used (Equation S5) to fit data obtained from the UVC-deionised water treatments:

$$Log_{10}N = Log_{10}((10^{Log_{10}N_0} - 10^{Log_{10}N_{res}}))\ \times 10^{\left(-\frac{d}{\delta}\right)^p} + 10^{Log_{10}N_{res}} \quad \text{(S5)}$$

Where N_0 and N were the bacterial colony forming units per mL of suspension (cfu/mL), before (influent) and after treatment (effluent), respectively, d is the UVC fluence (KJ/m^2)/applied dose, with δ and p being the scale and shape parameters for the data distribution, respectively, and N_{res} is the residual population resisting the applied treatment (Log_{10} CFU/mL). δ provides the fluence for the first decimal reduction (KJ/m^2), whereas the shape parameter p describes the downward (≥ 1) or upward (≤ 1) concavity of the curve (unitless).

Fitting any form of Weibull function to the UVC-treated borehole water data was not possible, since: a) only four fluences and/or contact times were attempted for testing the UVC inactivation efficiency, b) the natural bacterial load of the borehole water was below 4 Log_{10}, and c) scatter plots of Log_{10} inactivation versus fluence lacked a tailing phase. Thus, the Log_{10} linear model [38] was used to fit the data (Equation S6).

$$Log_{10}N = Log_{10}N_0 - k_{max}d \quad \text{(S6)}$$

Where k_{max} is the maximum inactivation rate constant, derived from the slope of the Log_{10} linear curve. To obtain the D value (equivalent to the δ parameter, i.e., the dose required to achieve the first decimal reduction) Equation S7 was used:

$$D = \frac{Ln10}{k_{max}} = \frac{2.303}{k_{max}} \quad \text{(S7)}$$

Text S.4.2 chemical disinfection; the disinfectant concentration decayed with pseudo-first order characteristics, and decay rate constants (k_d; (min^{-1})) were determined using Equation S8.

$$Ln\left(\frac{C_{disinfectant}}{C_{disinfectant,0}}\right) = -k_d t \quad \text{(S8)}$$

At any time point, disinfectant exposures were derived using Equation S9.

$$Disinfectant\ exposure = \int_0^t C_{disinfectant} dt \quad \text{(S9)}$$

Where $C_{disinfectant}$, is the concentration of available residual at time t.

Since the disinfectant's concentration varied during the inactivation process, a modified version of Hom's model (Haas and Joffe, 1984, cited in [39]) was used for the derivatisation of the kinetic parameters (Equation S10).

$$Ln\frac{N}{N_0} = -k_{disinfectantion} C_{disinfectant}{}^n t^m \left(\frac{1-exp\left(\frac{-k_d t}{m}\right)}{\frac{k_d t}{m}}\right) m \quad \text{(S10)}$$

Where $k_{disinfection}$, is Hom's inactivation rate constant ($mg^{-n}L^{n}min^{-m}$), n (usually close to unity) is the coefficient of dilution and m is the Hom's time exponent. The values of both exponents depend on the experimental conditions (temperature, pH, salinity, physicochemical properties of the treated water) and the characteristics of the microbe subjected to the treatment [40]. When the coefficient of dilution n > 1, the influence of the concentration factor in the bacterial inactivation is stronger than the length of the exposure [41]. Time exponent m is the distribution shape factor. When m > 1, the curve exhibits downward concavity with accumulated damage weakening the possibly resistant populations. When m < 1, the curves exhibit upward concavity, leading to tailing phenomena indicative of emergence of resilience to the treatment [40].

Having determined the k_{decay} rate constants from Equation S8, the m and n coefficients were calculated from the solution of Equation S10, to achieve the minimisation of the mean sum of squared error (MSE). Based on the Hom's modified model, the m and n coefficients would give rise to fractional reaction kinetic orders, reflecting the complexity of the disinfectant chemistry. 1.1 mg/L Cl_2 exhibited the lowest m coefficients in comparison to 0.75 mg/L ClO_2 and NaClO, reflecting its more complex side-reaction chemistry [42]. Since the bacterial inactivations exhibited different reaction orders, direct comparison of the disinfection rate constants was not possible.

Text S5. Assessing the goodness of model fitting

The GinaFit parameters, mean squared error (MSE), root mean sum of squared error (RMSE) and the adjusted R^2 (R^2 adj.), were used to assess the goodness of fit of the Weibull + tail model [36] to the data. R^2 adj. values >90% and RMSE values in the 0.3-0.7 range are reported. To assess over- or under-fitting, RMSE values were compared to the standard deviation of the residuals (STDEV). RMSE>>STDEV implied the use of a flexible model, with overfitting emerging from low signal to noise ratio. In contrast, for RMSE<<STDEV the model is considered too inflexible to capture the trend. The RMSE values derived from curve fitting to the UVC-inactivation data (Table 2) were only slightly different to the corresponding SD values, suggesting a reasonable description of the kinetic behaviour.

For curve fitting using GraphPad, the Sy.X (standard error of residual) parameter is reported. Sy.x is equivalent to RMSE. Both RMSE and Sy.X parameters require determination of the MSE, but whereas for RMSE estimation, (n-1) is required to modify the residual number,

for Sy.X, (n-k) is used (where k is the number of parameters, derived from the applied equation). Thus, Sy.x = RMSE, when a single parameter is used.

Text S6. HC-mediated bacterial inactivation

The *E. coli* Log_{10} (N_0/N) values achieved by HC alone ranged from 0.78 ± 0.06—0.84 ± 0.09 and from 0.62 ± 0.09—0.74 ± 0.03 for the T-sonic OM and PW hydrocavitators, respectively (Figure S3a). Log_{10} (N_0/N) differences between passages and cavitator type were not statistically significant ($P > 0.220$, n = 3). In comparison to UVC, the HC-mediated disinfection of *E. coli*-spiked deionised water was at least 7-fold weaker ($P < 0.0001$, n = 3) (Figure S3a).

The HC-mediated disinfection of *E. faecalis*-spiked deionised water (delivered to the cavitators at 15.0 L/min), revealed Log_{10} (N_0/N) values of 1.5 ± 0.1—1.6 ± 0.1 and of 1.30 ± 0.1—1.7 ± 0.2 cfu/mL for the T-sonic OM and PW cavitators, respectively (Figure S3b). For the T-Sonic OM there were no differences in the inactivations achieved between passages ($P > 0.460$, for n = 3), whereas for the T-Sonic PW, the first passage appeared more effective than consecutive passages ($P = 0.0327$, n = 3 (3rd), and $P = 0.0314$ n = 3 (5th)) (Figure S3b). The T-Sonic OM appeared more effective over the PW for the 3rd passage ($P = 0.0280$, n = 3) (Figure S3b). Overall, *E. faecalis* HC-based inactivations were at least 4-fold weaker than those of UVC ($P = 0.0002$, n = 3), but at least 1.9-fold higher ($P = 0.0216$, n = 3) than those of *E. coli* (Figure S3b), suggesting that HC was more effective in reducing the *E. faecalis* than the *E. coli* load of deionised water. Consequently, we opted for using only the OM cavitator in subsequent bench-scale UVC/HC studies because of its moderately higher microbial inactivation propensity than the PW cavitator.

As highlighted in section 4.1 (Main Text) and demonstrated in Figure 5, cavitator OM-mediated ORP changes were apparent in the hybrid UVC/HC treatments of *E. faecalis* spiked deionised water. However, analysis of indicator parameters of tap-water effluents collected before and after individual HC treatments with both cavitators, revealed statistically significant effects only for the PW cavitator. By the 5th passage there was a nearly 9% decrease in the concentration of TDS, which coincided with a similar percentage reduction in conductivity (Main Text Figure 6), consistent with the elimination of organic/inorganic matter. There was at least an 8% reduction in pH and a maximum of 39% increase in ORP, indicative of mild acidification of water and the generation of reactive oxygen species (Main Text Figure 6).

Thus, due to the performance, and the capacity of the PW cavitator to operate at higher flow rates than the OM cavitator, PW was implemented in preliminary plant pilot-work.

Text S7. Total standard deviation calculations from individual group variances

For determining whether the bacterial inactivation achieved by the UVC/HC hybrid treatments were the result of additive or synergistic effects of the individual disinfection technologies, the mean Log_{10} reductions achieved by the UVC and the hydrocavitation technologies individually were added together and compared to the Log_{10} reductions achieved by the combined treatments. The total standard deviation for the Log_{10} inactivation of the UVC + hydrocavitation combination was calculated from the population variances of the mean Log_{10} inactivation achieved by the individual technologies, using Equation S11.

$$Total\ Standard\ Deviation, \sigma_{total} = \sqrt{{\sigma_{UVC}}^2 + {\sigma_{HC}}^2} \quad \text{(S11)}$$

Where, ${\sigma_{UVC}}^2$ and ${\sigma_{HC}}^2$ are the population variances of the Log_{10} inactivations achieved in individual UVC and HC triplicate experiments, respectively. With the total standard deviation calculated, t-test comparisons (95% confidence intervals, significance; $P < 0.05$) between UVC and hybrid treatments (UVC-HC, HC-UVC) were conducted in GraphPad Prism.

Text S8. ClO_2 decomposition half-lives

ClO_2 gas in aqueous solutions, in a concentration and pH dependent manner, rapidly decomposes to generate chlorate, chlorite, chlorite and oxygen [47]. Even dilute ClO_2 solutions (provided they are protected from light exposure) have been reported to exhibit short half-lives (15-180 minutes; [48, 49]). Alkalinity encourages the faster decomposition of ClO_2 [50], with the chlorite:chlorate production ratio increasing with decreasing [ClO_2] in the micromolar range. Since the recommended values for chlorite and chlorate, on ClO_2 disinfection are 0.7 mg/mL, with the possibility of being exceeded following ClO_2-treatments, ClO_2-mediated borehole water inactivation was attempted at 0.75 and 0.3 mg/L. In RO water, ClO_2 exhibited half-lives of ~3-6 hr (Figure S4a). In agreement with previous studies [51], alkalinity encouraged ClO_2 decomposition (Figure S4b), yielding half-lives of 0.18 and 0.20 hours (10.8 and 12 min) for 0.75 and 0.3 mg/L ClO_2, respectively (Table S9).

Text S9. Chemical demand

Because of its lack of reactivity with NH_4^+ and Br^-, ClO_2 does not display the classical chlorination demand curves, with [free chlorine] proportionally rising after the chemical demand is satisfied and the break-point chlorination is reached [53]. However, ClO_2 has been shown to rapidly react with I^-, CN^-, NO_2^-, SO_3^{2-}, Fe^{2+}, Mn^{2+}, aliphatic tertiary amines and heterocyclic nitrogenous compounds [54]. Consequently, there is an apparent ClO_2 demand that needs to be met for observing effective biocidal action.

For the chosen ClO_2 doses in RO water, proportionality relationships between applied and total ClO_2, 0.2-2 hr post-dosing, respectively, suggested the lack of chemical demand (Figure S6a). However, applied ClO_2 doses in borehole water, shifted the proportionality relationship to the right of the origin [55], implying that a 0.06 mg/L chemical demand had been met with the applied treatments (Figure S6b).

Unlike ClO_2, RO water-dosing with ≤ 0.65 mg/L NaClO was not directly proportional to the free oxidant, suggesting the presence of metals, organics and ammonia, interacting with the applied NaClO to build combined residual [56]. RO water-dosing with NaClO ≥ 0.65 mg/L appeared to meet the chemical demand with free residual proportionally raising with increasing [NaClO] (Figure S7a). However, the chemical demand of borehole water appeared higher, with at least 0.75 ppm NaClO required to reach the break-point chlorination (Figure S7a).

Regarding the gas chlorination of blended water, the Naxxar blend (70% (v/v) RO and 30% (v/v) borehole water, pH 8.0) exhibited a curve with a breakpoint at 0.55 mg/L chlorine, whereas the Ta' Qali blend's curve revealed a breakpoint of 0.85 mg/L (Figure S7b). Thus, for both *ex* and *in situ* chlorination the chemical demand raised by the different water qualities was in the 0.55-0.85 mg/L range.

Text S10. Chemical analyses of borehole water samples following the chemical inactivations

The natural organic matter (NOM) of borehole water, as well as its nitrogenous inorganic content, and the possible herbicide, pesticide, fungicide and industrial effluent pollutants that can alter its chemical make-up, will determine the type of DBPs formed following the disinfection treatment. Nitrogenous-DBPs [57], THMs and THAs, even when at concentrations below the guidance values proposed to ensure the consumer's health and safety, can impact on the water's sensory characteristics. With pH, temperature and chemical

composition of borehole water dictating the quality and quantity of the DBPs produced on disinfection, seasonal monitoring of the water's content becomes essential, particularly when considering the application of electrochlorination in the treatment of borehole water.

On generation, NaClO will dissociate to Na^+ and ClO^-. On NaClO dosing, ClO^- will generate HClO accompanied with an increase in borehole water's pH (Figure S10). At pH range of 5-9 both HClO and ClO- are present. Hypochlorite will disproportionate to generate chlorate and chlorite with a subsequent decrease in pH, in a reaction that is dependent on the [HClO]/[ClO^-] ratio and will generate chlorate with decreasing pH [58]. It appears that within 30 min and for the 0.75 mg/L dose, the [HClO]/[ClO^-] ratio is sufficiently high to generate chlorate, detectable by 2.5 hours (Main Text Table 5, Figure S10).

At pH 8.4, monochloramine becomes the major chloramine present, offering residual disinfection without compromising the organoleptic properties of water (no odour sensory thresholds have been reported for monochloramine, whereas taste and odour thresholds for dichloramine (pH 4-6 or following the auto-decomposition of monochloramine over pH 6-8.5) were 0.48 and 0.65 mg/L respectively [22, 59]. Thus, NaClO can potentially offer additional biocidal activity relative to ClO_2.

In contrast to NaClO disinfection, whose DBP repertoire resembles that of gaseous chlorine, ClO_2 decay can generate two main by-products with exposure to UV/sunlight [17] and alkalinity [51] encouraging ClO_2 decomposition (Main Text Table 5).

Table S1. Administered UVC irradiation doses for the tested flow rates and minimum exposure times (t_{RMin}) for laminar flows.

***E. coli* flow rates (L/min)**	**t_{RMin} (Min)**	**t_{RMin} dose (kJ/m^2)**	**3 x t_{RMin} (Min)**	**3 x t_{RMin} dose (kJ/m^2)**	**5 x t_{RMin} (Min)**	**5 x t_{RMin} dose (kJ/m^2)**
0.17	15.2	19.2	45.5	57.7	75.8	96.1
0.40	6.44	13.0	19.3	39.0	32.2	65.0
0.60	4.29	7.30	12.9	21.9	21.4	36.5
1.13	2.28	4.20	6.84	12.6	11.4	21.0
2.50	1.03	2.40	3.09	7.20	5.15	12.0
5.50	0.47	0.85	1.41	2.55	2.35	4.25
9.50	0.27	0.35	0.81	1.05	1.35	1.75
***E. faecalis* flow rates (L/min)**	**t_{RMin} (Min)**	**t_{RMin} dose (kJ/m^2)**	**3 x t_{RMin} (Min)**	**3 x t_{RMin} dose (kJ/m^2)**	**5 x t_{RMin} (Min)**	**5 x t_{RMin} dose (kJ/m^2)**
0.083	31.0	45.3	93.1	223	155	371
0.27	9.54	13.9	28.6	68.5	47.7	114
0.86	3.00	6.70	9.00	21.1	15.0	35.9
1.70	1.52	3.16	4.56	9.14	7.60	18.1

3.00	0.86	1.42	2.58	4.27	4.30	7.50
10.5	0.25	0.36	0.75	1.29	1.25	1.91
15.0	0.17	0.25	0.51	0.78	0.85	1.34

Table S2. A comparison of CAPEX and OPEX costs[1] for the implementation of UVC, ClO_2, and electrochlorination (NaClO) in groundwater (borehole) treatments.

Disinfection Technology			**UVC**	**ClO_2 (4.2 g/h required)**	**Electrochlorination (NaClO) (11.2 g/h required)**
CAPEX (€)	Equipment		3,841.44	13,623.43	16,785.68
	Technical Assistance: Installation, Commissioning, on-site training		2,000.00	2,000.00	2,000.00
Total CAPEX (€)			5,841.44	15,623.43	18,785.68
OPEX (€)	Labour		195.84	195.84	195.84
	Parts p.a. (in-stock)		1,620.20	11,570.00	11,570.00
	Raw Material Consumption	Energy (€/kWh)	0.0230	0.0024	0.0765
		Process Consumables	150.00 p.a.	0.43	0.04
	Service (per annum)		1,850.00	5,414.00	5,414.00
OPEX/m^3 water produced (€)			0.0521	0.1906	0.1680

[1]Mean groundwater flow rate (peak season) was taken as 14 m^3/hr (Main Text section 2.9).

Table S3. A comparison of CAPEX and OPEX costs[1] for the implementation of UVC, ClO_2, and electrochlorination (NaClO) in desalinated water (RO) treatments.

Disinfection Technology		**UVC**	**ClO_2 (806.88 g/h required)**	**Electrochlorination (NaClO) (2151.68 g/h required)**
CAPEX (€)	Equipment	478,553.94	260,379.00	535,885.74
	Technical Assistance: Installation, Commissioning, on-site training	2,000.00	2,000.00	2,000.00
Total CAPEX (€)		480,553.94	262,379.00	537,885.74
OPEX (€)	Labour	5,287.68	587.52	3,525.12
	Parts p.a. (in-stock)	43,745.40	34,710.00	208,260.00
	Raw Material Consumption: Energy (€/kWh)	2.41	0.92	4.02
	Raw Material Consumption: Process Consumables	4,050.00	83.35	7.46
	Service (per annum)	1,850.00	5,414.00	5,414.00
OPEX /m^3 water produced (€)		0.0059	0.0334	0.0153

[1]Mean desalination flow rate (peak season) was taken as 2689.6 m^3/h /hr (Main Text section 2.9). To match the peak flow rate, costs were determined for 27 UVC-units, 3 ClO_2 units, and 18 electrochlorinator units.

Table S4. A comparison of CAPEX and OPEX costs[1] for the implementation of UVC, ClO_2, and electrochlorination (NaClO) in reservoir water (blended water) treatments.

Disinfection Technology		**UVC**	**ClO_2 (904.04 g/h required)**	**Electrochlorination (NaClO) (2410.77 g/h required)**
CAPEX (€)	Equipment	567,175.04	347,172.00	654,971.46
	Technical Assistance: Installation, Commissioning, on-site training	2,000.00	2,000.00	2,000.00
Total CAPEX (€)		569,175.04	349,172.00	656,971.46
OPEX (€)	Labour (monthly)	6,266.88	783.36	4,308.48
	Parts p.a. (in-stock)	51,846.40	46,280.00	254,540.00
	Raw Material Consumption: Energy (€/kWh)	2.86	1.22	4.91
	Raw Material Consumption: Process Consumables	4,800.00	93.39	8.36
	Service (per annum)	1,850.00	5,414.00	5,414.00
OPEX/m^3 water produced (€)		0.0063	0.0338	0.0164

[1]Mean reservoir water flow rate (peak season) was taken as 3013.46 m^3/h /hr (Main Text section 2.9). To match the peak flow rate, costs were determined for 32 UVC-units, 4 ClO_2 units, and 22 electrochlorinator units.

Table S5. Scoring system for assessing the microbial inactivation efficiency of UVC.

	RO water (pH 7.5)							
	***E. faecalis* inactivation**		***E. coli* inactivation**		**Stability**	**Minimum Log_{10} inactivation of 2**	**Overall residual activity[1]**	**Total score**
	4D	$Log_{10}(N_{res})$ (cfu/mL)	4D	$Log_{10}(N_{res})$ (cfu/mL)				
UVC	1	1	2	2	2	2	0	10
	Borehole water (pH 8.4)							
	***E. faecalis* inactivation**		**Total Bacterial inactivation**		**Stability**	**Minimum Log_{10} inactivation of 2**	**Overall residual activity**	**Total score**
	4D[†]	$Log_{10}(N_{res})$ (cfu/mL)[2]	4D	$Log_{10}(N_{res})$ (cfu/mL)				
UVC	1	1	1	-	2	2	0	7
	Combined performance (summative scores)							
UVC	17							

[1]Residual activity points are allocated to the technology based on the chemical species expected to carry out secondary disinfection. UVC offers no residual activity and thus scores 0. In contrast, HC can generate reactive oxygen species and as such scores 1, since chemical disinfection technologies, i.e., gas chlorination and *in situ* NaClO production generate more DBPs that can contribute to microbial inactivation and can score 2-3 points.

[2]Assumed behaviour based on the UVC inactivation performance in RO water. Since *E. faecalis* was more resistant to the UVC treatment in RO water than *E. coli*, it is unlikely it would exhibit higher sensitivity in borehole water where the total carbon content is higher, and the microbial flora of the ground water can contribute to biofilm formation.

Table S6. Scoring system for assessing the microbial inactivation efficiencies[1] of the alternative-to-gas chlorination technologies.

	RO water (pH 7.5)							
	***E. faecalis* inactivation**		***E. coli* inactivation**		**Stability**	**Minimum Log_{10} inactivation of 2**	**Overall residual activity[2]**	**Total score**
	Rate constant	4 Log_{10} time	Rate constant	4 Log_{10} time				
ClO_2	1	1	1	1	1	2	2	9
NaClO	2	2	2	2	2	2	3	15
	Borehole water (pH 8.4)							
	***E. faecalis* inactivation**		***E. coli* inactivation**		**Stability**	**Minimum Log_{10} inactivation of 2**	**Overall residual activity**	**Total score**
	Rate constant	4 Log_{10} time	Rate constant	4 Log_{10} time				
ClO_2	2	2	2	-	1	2	2	11
NaClO	1	1	2	-	2	2	3	11
	Combined performance (summative scores)							
ClO_2	20							
NaClO	26							

[1]Assessments were based on the experimental observations of this study. 2 marks were allocated to the best performing technology and 1 point to the second ranking one. As both technologies achieved a minimum 2 Log_{10} reduction, they were allocated 2 points each.

[2]Residual activity points are allocated to the technology based on the chemical species expected to carry out secondary disinfection. *Gas chlorination* and *in situ* NaClO generation have more complex chemistry than chlorine dioxide, and several of their DBPs can contribute to microbial inactivation. As such, 2 points were allocated to NaClO as opposed to 1 point allocated for ClO_2.

1

Table S7. Provisions for implementation of alternative-to-gas chlorination disinfection technologies in the current settings.

	UVC		ClO_2		NaClO	
Question	**Provisions**	**Justification**	**Provisions**	**Justification**	**Provisions**	**Justification**
1.1	Hardness control	To prevent deposition of $CaCO_3$ on sleeve and reduction of UVC intensity	Suitable pH	To reduce chlorite and chlorate content [17]	Suitable pH	For control of hypochlorous acid and hypochlorite balance [18]
	UVC lamp sleeve maintenance	To encourage full UVC intensity reaching the bacterial load	Suitable temperature	Lower T, higher [chlorate]. Higher T, higher [chlorite] [17]	Suitable temperature	Lower T, higher [hypochlorite]. Higher T, higher [hypochlorous acid] [18]
	UVC sensor maintenance	To encourage full UVC intensity reaching the bacterial load	Chlorate/chlorite monitoring	Reduction of DBP formation	Chlorate/chlorite monitoring	Reduction of DBP formation
	Biofilm formation control	Deposition of biofilm on sleeve will reduce UVC intensity	Free chlorine monitoring	Reduction of THM formation	Free chlorine monitoring	Reduction of THM formation
	Appropriate flow rate	For optimal contact times/doses	Ammonia monitoring (particularly at borehole level)	For chloramine control	Ammonia monitoring (particularly at borehole level)	For chloramine control
	Spatial requirements for instrumentation	For chloramine control	Photolysis prevention	For decomposition prevention and avoidance of chlorate/chlorite and DBP formation [19]	Trace metal monitoring	For decomposition prevention and avoidance of chlorate/chlorite and DBP formation [20]
	Turbidity control	To encourage full UVC intensity reaching the bacterial load	Monitoring of phenols	For free chlorine and DBP formation [21]	Monitoring of organics	For prevention of chloroform/bromoform formation [22]

	Monitoring Ammonia	For predicting nitrate content	Pipe maintenance	Decomposition of plastic and metal [23]	Pipe maintenance	Requirement for PVC, CPVC pipes [24]
	Monitoring Nitrates/nitrites	To discourage nitrite formation	TOC/DOC monitoring	Chlorite to chlorate transformation in the presence of organics [17]	TOC/DOC monitoring	For control of DBP formation [25]
	Monitoring TOC/DOC	Disintegration of quaternary ammonium compounds may generate ammonia and chloride	Bromoform monitoring	Higher [ClO_2], lower risk for Br-DBP formation [26]	Bromoform monitoring	Bromide oxidation to bromate [27]
					HAA monitoring	For reduction of toxic disinfection by-products
2.1			Staff training	For smooth operational practices	Staff training	For smooth operational practices
2.2	Suitable technical support	For effective installation and servicing	Spatial requirements	Instrumentation size dictates the most appropriate installation site	Suitable technical support	For effective installation and servicing
	Pump(s)	For delivery of influent	Vent-pipes/gas release	For occupational safe practices	Spatial requirements	Instrumentation size dictates the most appropriate installation site
	Spatial requirements	Instrumentation size dictates the most appropriate installation site	Pump(s)	For delivery of influent	Storage of NaClO	Light protection before dosing the influent
					Dosing Pump	For injection of NaClO
					Pump(s)	For delivery of influent
2.3	Staff training	For smooth operational practices	Staff training	For smooth operational practices	Staff training	For smooth operational practices
			Installation requirements	For effective decontamination	Installation requirements	For effective decontamination
			Gas sensor	For occupational safe practices	Capacity for H_2 gas release	For occupational safe practices

4.1	UVC instrumentation must fit the current set-ups	To reduce cost and environmental impacts				
4.3	Applicability of all implementation requirements	As previously justified	Applicability of all implementation requirements	As previously justified	Applicability of all implementation requirements	As previously justified
	Co-administer with ClO_2	To encourage reduction of and control DBPs	Co-administer with UVC	To reduce the risk of ClO_2 photolysis and prevent chlorite/chlorate formation while encouraging DBP elimination [15]	Must succeed UVC	To reduce NaClO dosing and encourage higher coliform decontamination [28]
					Must precede ClO_2	To encourage reduction of THMs and reduce the effective ClO_2 dose [29]
	Use after *ex situ*/*in situ* chlorination	To encourage disinfectant decay [30] and reduce the generation of organic precursors of DBPs (inclusive of chloramines) [15]	ClO_2 monitoring	To comply with the EU directive guidance values for sum of ClO_2, chlorite and chlorate	Regular monitoring of demand curves	To make informed decisions on dosing relative to chemical, oxidant, and microbiological demand

Table S8. Assessment of disinfection technologies in terms of greenhouse gas emissions (CO_2 equivalents).

Disinfection technology instrument	**Type**	**Power (W)**	**Energy consumption (kWh)**	**Kg CO_2 emissions per kWh (USA)[1]**	**Kg CO_2 emissions per kWh (Malta)[2]**
Mini-chlorgen-Lutz (NaClO) +	30	30	0.15	0.0579	0.05865
Dosing pump	6	28	0.028	0.0108	0.010948
Lotus mini-Emec (ClO_2)	8	50	0.05	0.0193	0.01955
UVC-Sita	405	38	0.03	0.01158	0.01173

[1] Estimations with USA average factor for 2020 0.386 kgCO_2e/kWh.

[2]Estimations with Malta's emissions factor for 2022; 0.391 kgCO_2e/kWh (Carbon Footprint Ltd).

Table S9. Determined ClO_2 and NaClO half-lives in RO and borehole waters at 19.4°C.

	RO water (pH 7.5)			**Borehole water (pH 8.4)**		
[ClO_2] (mg/L)	**Half-life[1] (hr)**	**R^2**	**Sy.x[2]**	**Half-life (hr)**	**R^2**	**Sy.x**
15.0	3.5 ± 0.3	0.9857	0.6689	n.d.[3]	n.d.	n.d.
7.50	3.4 ± 0.3	0.9837	0.3792	n.d.	n.d.	n.d.
3.80	3.1 ± 0.4	0.9739	0.2496	n.d.	n.d.	n.d.
1.90	3.1 ± 0.5	0.9419	0.1705	n.d.	n.d.	n.d.
0.90	4.4 ± 0.5	0.9756	0.0546	n.d.	n.d.	n.d.
0.75	3.4 ± 0.3	0.9760	0.0432	0.18 ± 0.03	0.9624	0.0497
0.30	6.0 ± 1.5	0.9277	0.0249	0.20 ± 0.03	0.9690	0.0188
[NaClO] (mg/L)	**Half-life (hr)**	**R^2**	**Sy.x**	**Half-life (hr)**	**R^2**	**Sy.x**
2.10	1.5 ± 0.6	0.9417	0.17520	n.d.	n.d.	n.d.
1.10	2.4 ± 0.3	0.9732	0.05895	n.d.	n.d.	n.d.
0.75	3.7 ± 0.3	0.9892	0.02413	3.2 ± 0.5	0.9611	0.0267
0.30	3.5 ± 1.0	0.8947	0.01884	3.4 ± 5.2	0.7258	0.0187
0.15	3.7 ± 1.1	0.8973	0.01271	n.d.	n.d.	n.d.

[1]Values are provided with their standard error for 95% confidence intervals.

[2]Sy.x is the standard deviation of the residuals, a value equivalent to RMSE. The lower its magnitude relative to Y, the better the goodness of fit.

[3]n.d. stands for not determined.

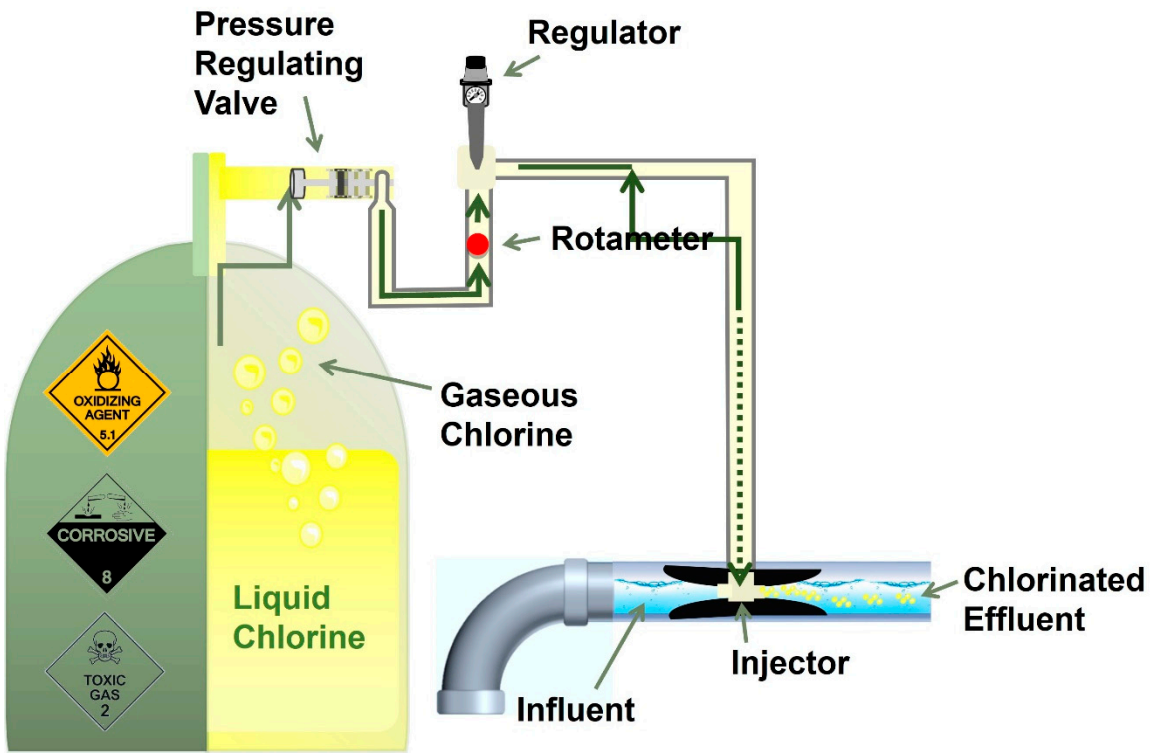

Figure S1. Cartoon of the chlorination set-up used in the disinfection of the Maltese potable water.

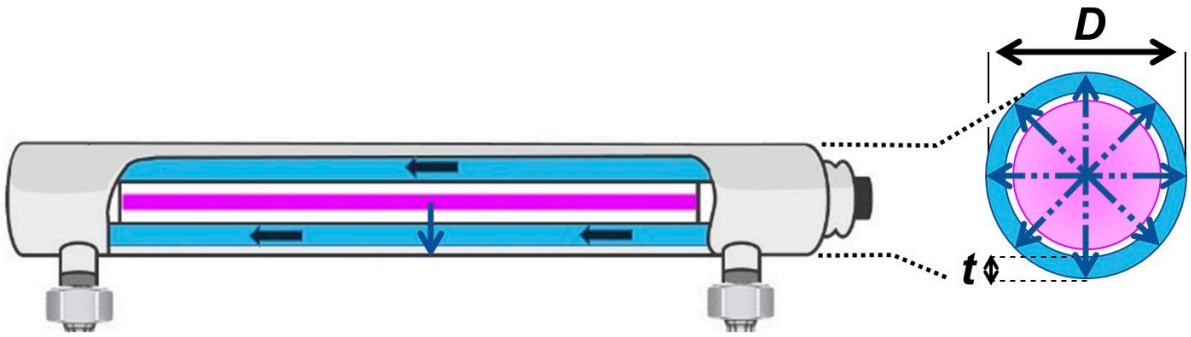

Figure S2. Cartoon of the UVC-405 sita lamp system with its cross-section. Water can flow in the blue shaded area surrounding the sleeved UVC lamp (dark blue arrows). For any tube the cross-sectional area calculation would require knowledge of its diameter (*D*) and its thickness (*t*). In this system there is a tube within a tube, i.e., a hollow cylindrical object. Hence, the distance from the sleeve of the lamp to the inner wall of the chamber would be the tube's thickness, which is also the dimension accessible to the water flow. For largely monochromatic light, radial distribution of irradiance is assumed. Note that dimensions are not to scale.

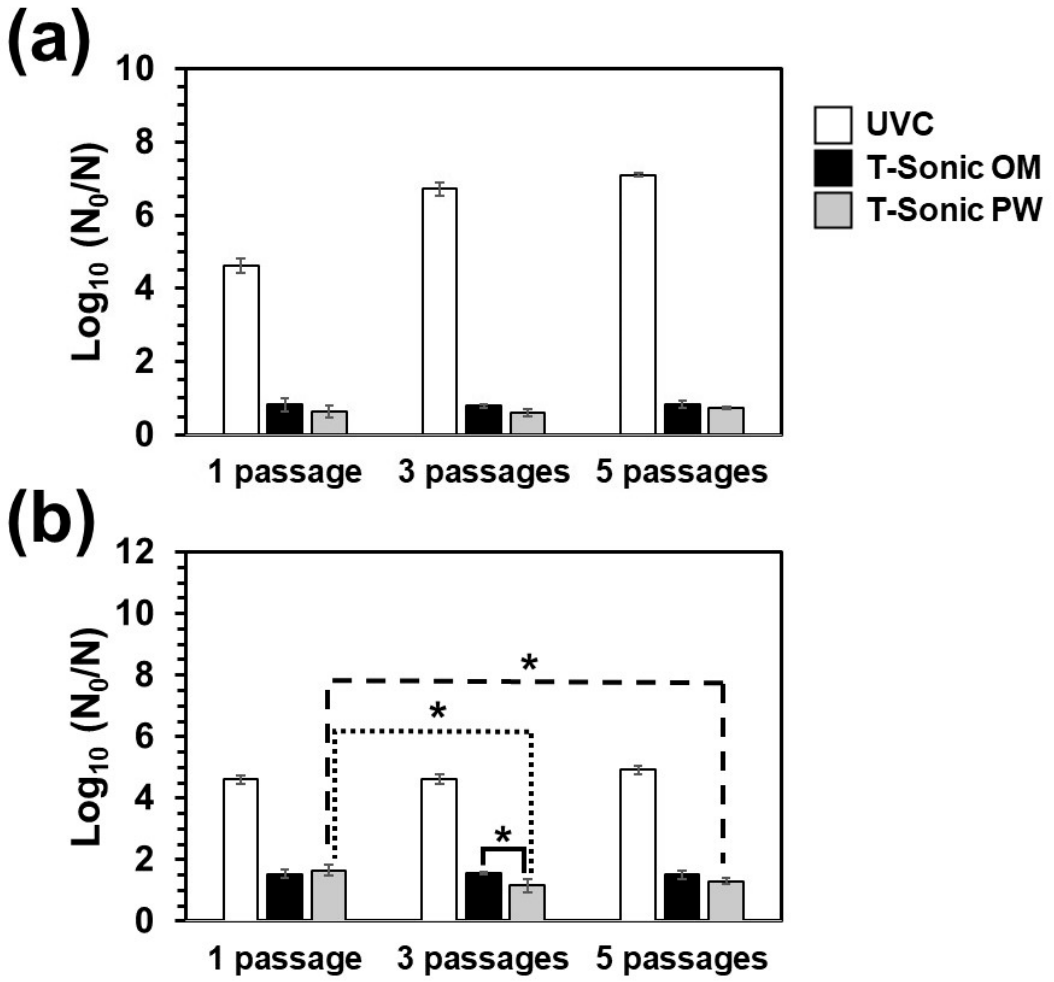

Figure S3. Microbial inactivation (Log_{10} (N_0/N) by hydrocavitation using the Treelium® T-Sonic OM and T-Sonic PW. (a) Inactivations in *E. coli*-spiked deionised water delivered to the cavitators at 9.5 L/min. (b) Inactivations in *E. faecalis*-spiked deionised water delivered to the cavitators at 15 L/min. The applied UVC doses for *E. coli* were 0.35 (passage 1), 1.05 (passage 3) and 1.75 (passage 5) kJ/m^2. The applied UVC doses for *E. faecalis* were 0.25 (passage 1), 0.78 (passage 3) and 1.34 (passage 5) kJ/m^2. For the 9.5 L/min flow, HC exposures were 0.016, 0.0048 and 0.08 min, for single, triple, and quintuple passages, respectively. For the 15 L/min flow HC exposures were 0.023, 0.069 and 0.115 min, for single, triple, and quintuple passages, respectively. Bars represent the standard error of triplicate measurements. Asterisks denote significance for 95% confidence for the indicated-by-bracket comparisons: solid lines (comparison between cavitators at 3rd passage), dotted line (comparison between 1st and 3rd passages for the T-Sonic PW cavitator), and dashed line (comparison between 1st and 5th passages for the T-Sonic PW cavitator). $P \leq 0.05$ (*), n = 3.

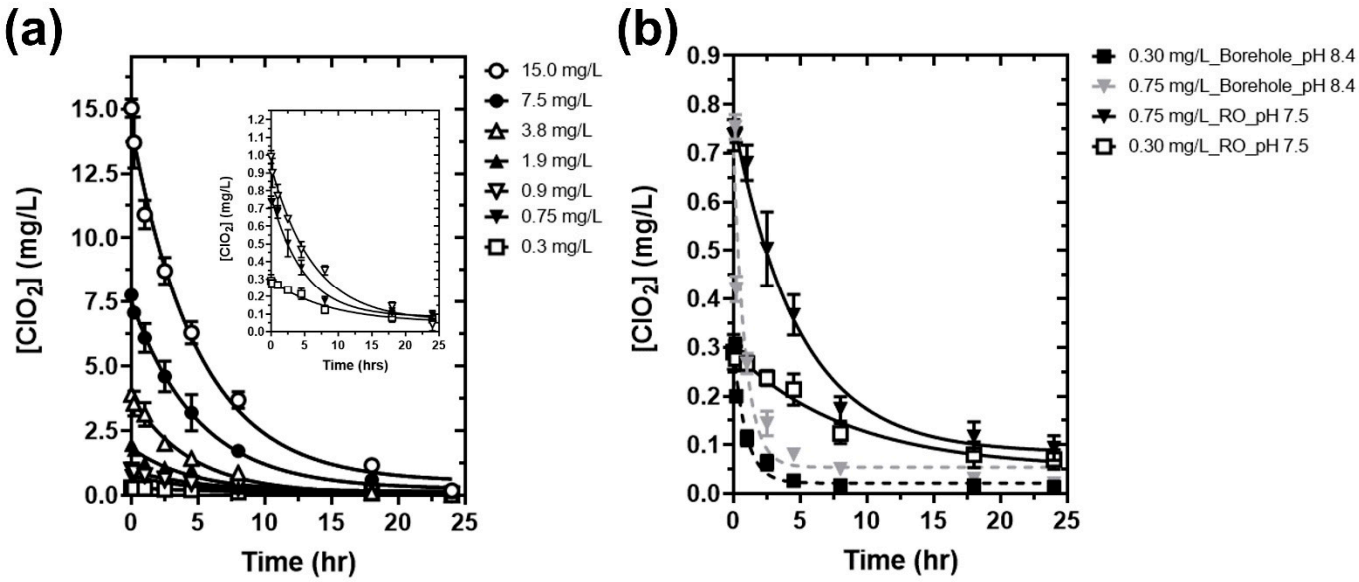

Figure S4. First-order ClO_2 decay fits on potable water dosed with different concentrations of ClO_2. (a) Concentration-dependent decompositions in RO water (pH 7.5) without biological demand. Inset shows the first-order decays of the three lowest doses for clarity. (b) Comparative evaluation of ClO_2 decomposition at 0.75 and 0.3 mg/L in RO (pH 7.5) and borehole (pH 8.4) waters in the absence of biological demand. Bars denote the standard deviation of triplicate measurements. ClO_2 concentrations were colourimetrically determined using the chlorophenol red indicator method (Main Text section 2.5).

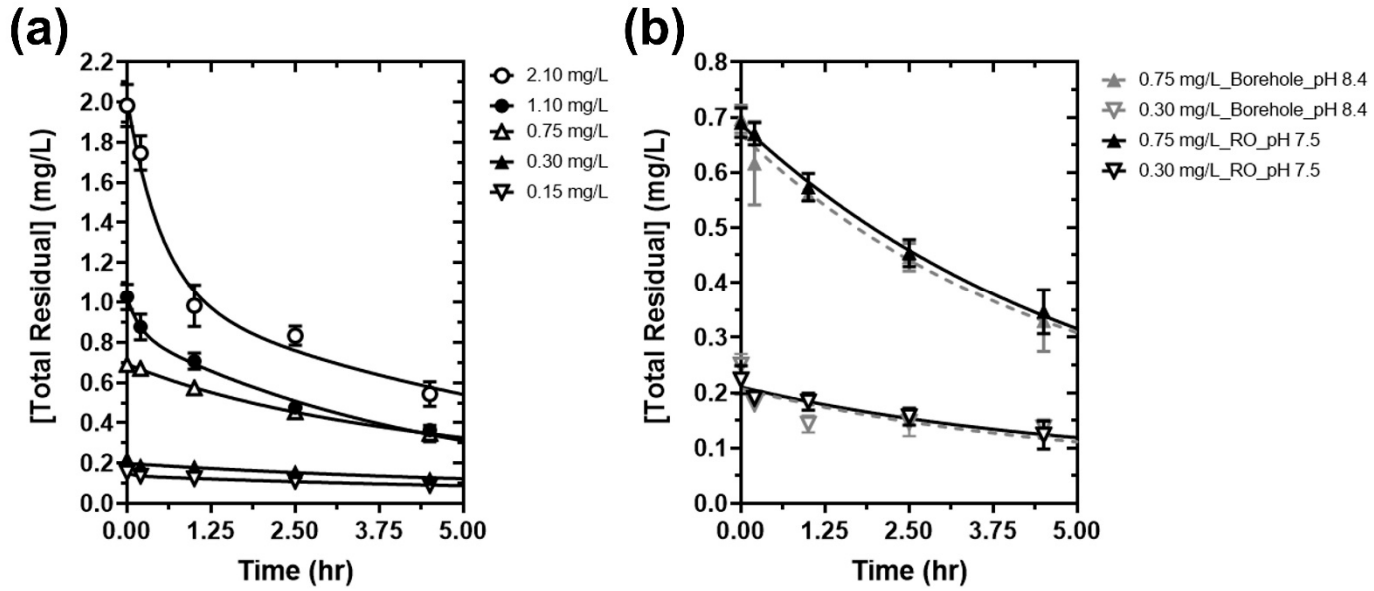

Figure S5. First-order NaClO decay fits on potable water dosed with different NaClO concentrations. (a) Concentration-dependent decompositions in RO water (pH 7.5) without biological demand. (b) Comparative evaluation of NaClO decomposition at 0.75 and 0.3 mg/L in RO (pH 7.5) and borehole (pH 8.4) waters in the absence of biological demand. Bars denote the standard deviation of triplicate measurements. Total and free chlorine were colourimetrically determined using the DPD-3 and DPD-1 indicator methods (main Text section 2.5).

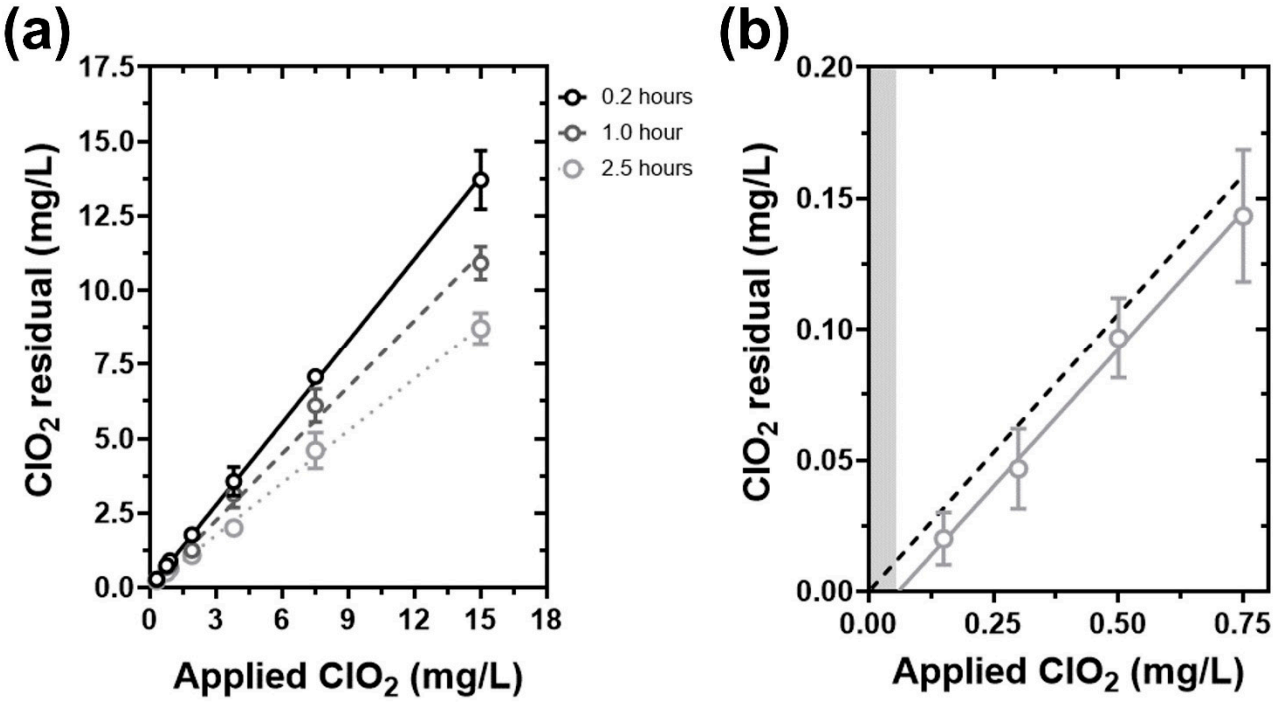

Figure S6. Dose and residual ClO_2 relationships in potable water. (a) Proportionality relationships in RO water with lines intercepting x = 0, y = 0 over different time points. (b) Proportionality relationship in borehole water at 2.5 hours, showing a shift along the x-axis relative to the dashed line that intercepts the origin (lack of demand). Bars denote the standard deviation of triplicate measurements. ClO_2 concentrations were colourimetrically determined using the chlorophenol red indicator method (main Text section 2.5).

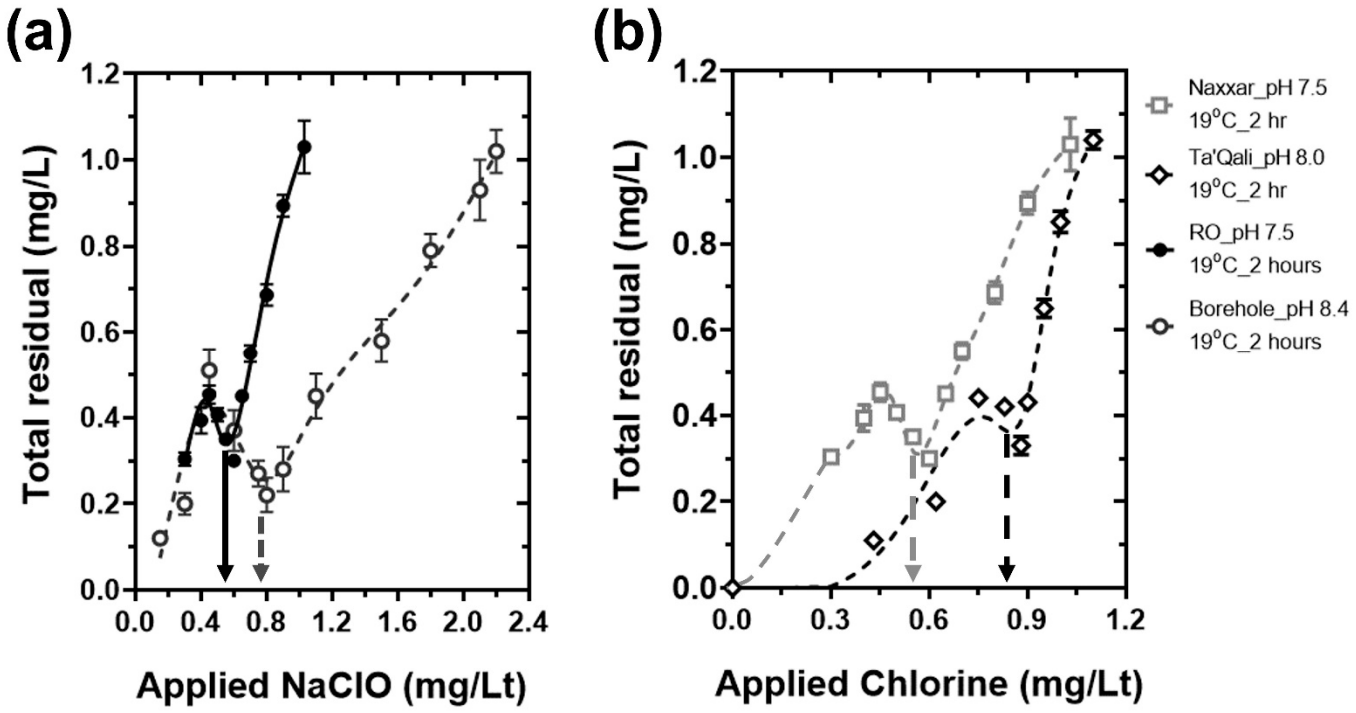

Figure S7. Breakpoint curves of NaClO- and chlorine-treated waters. (a) Changes in total residual with applied NaClO on borehole (pH 7.5) and RO (pH 8.4) waters. (b) Changes in total residual with applied chlorine dose on potable water blends (Naxxar (pH 7.5) and Ta' Qali (pH 8.0)). Investigations were conducted in the absence of biological demand. Bars denote the standard deviation of triplicate measurements. Arrows denote apparent chlorination breakpoints. Total and free chlorine were colourimetrically determined using the DPD-3 and DPD-1 indicator methods (main Text section 2.5).

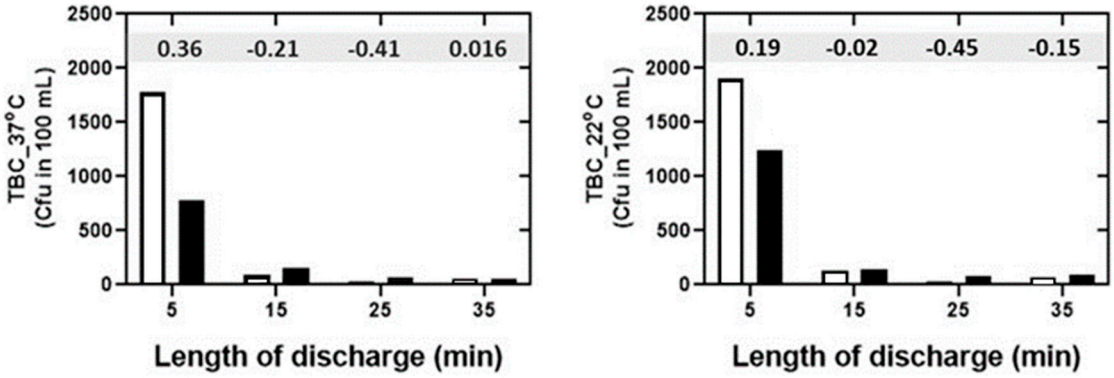

Figure S8. Dependence of HC inactivation efficiency on total bacterial count (TBC) input. 5 min following pipe-washout the total bacterial load input (whether at 37ºC or 22ºC) is high, and HC (inlet pressure 2 bar, outlet pressure 1 bar) achieves a minimum of 0.36 and 0.19 Log_{10} reductions (highlighted in light grey) for TBC at 37ºC and 22ºC, respectively. Extending the discharge time, lowers the input count significantly, negating disinfection and challenging the tested technology.

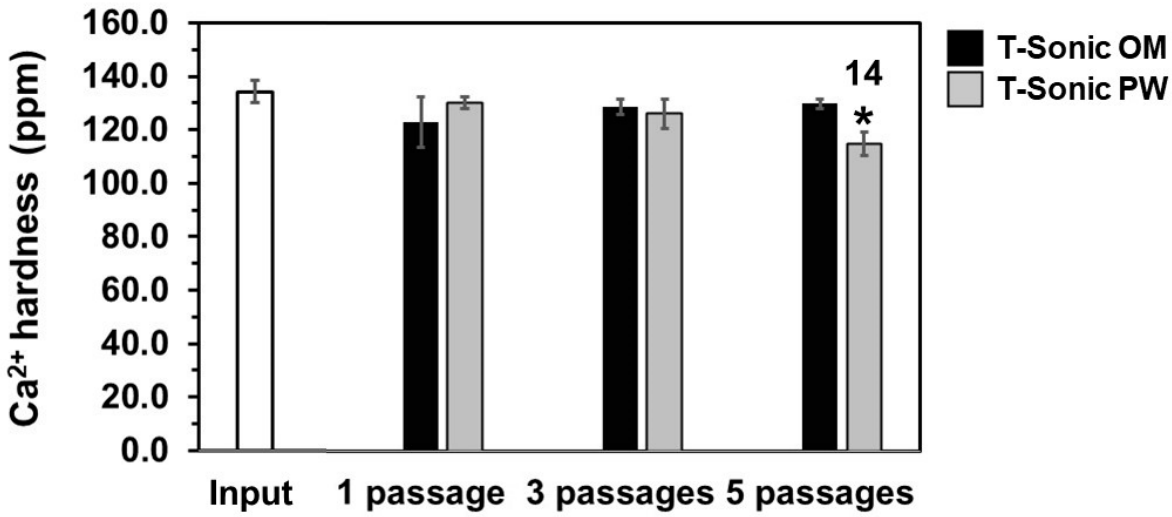

Figure S9. Ca^{2+} hardness measurements on *E. faecalis*-infected tap-water following treatments with T-Sonic OM and T-Sonic PW cavitators over different passages. Bars represent the standard deviation of triplicate measurements. Asterisks denote level of significance for 95% confidence for comparisons to the input: $P \leq 0.05$ (*), $P = 0.001$ - 0.01 (**), and $P \leq 0.001$ (***) for n = 3. Numbers above asterisks denote the percentage change relative to the input.

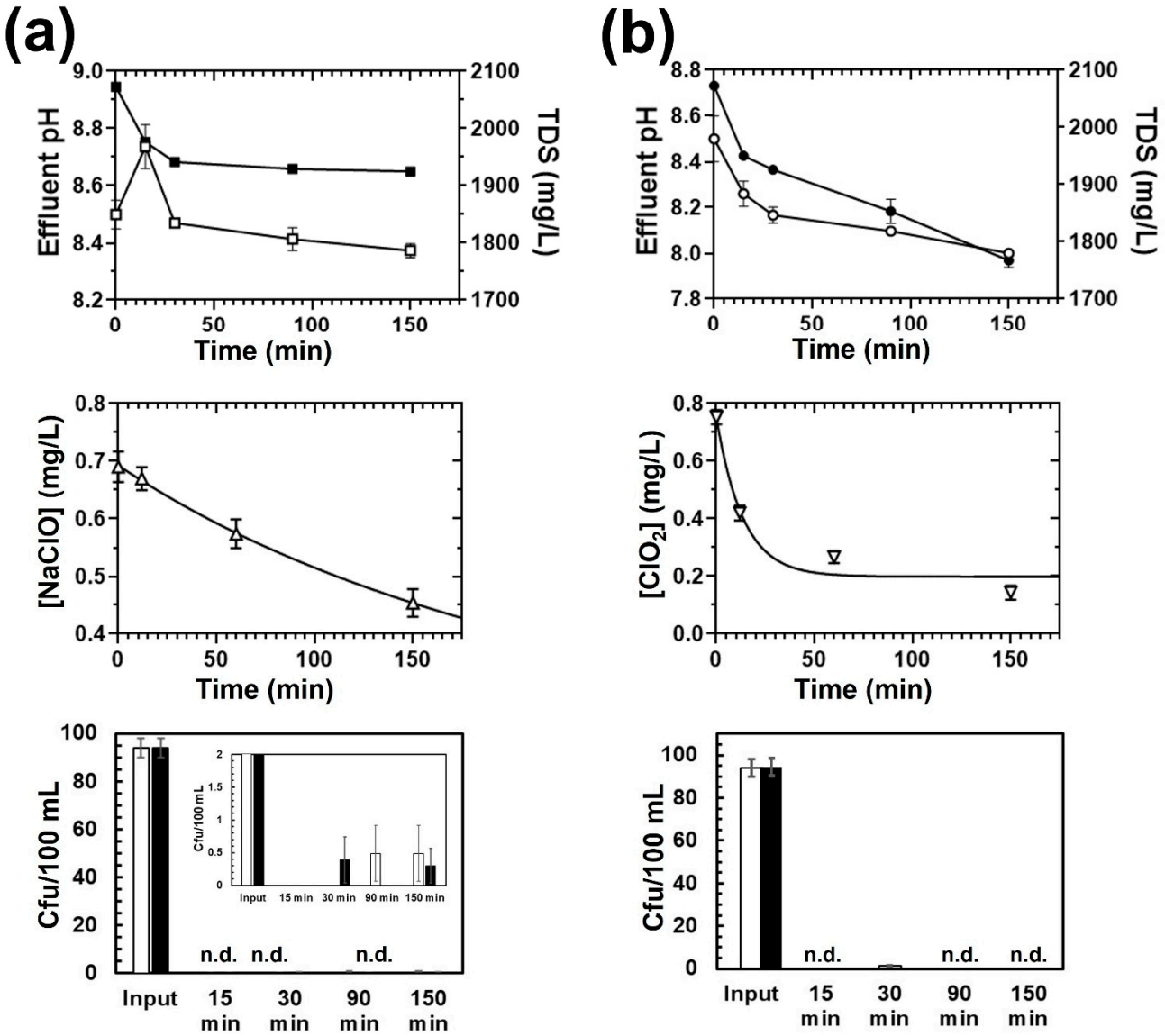

Figure S10. Time course of pH, TDS, concentration of residual and cfu/100 mL TBS (37ºC) in disinfected borehole water effluents. (a) Time course following NaClO-based disinfection. (b) Time course following the ClO_2-based disinfection. Top panels; pH and TDS changes with time. Middle panels; disinfectant decay. Bottom panels; TBC (37ºC) inactivation profiles with length of exposure. For 0.3 mg/L NaClO (white histograms), and for filtration volume of 250 mL, inactivation was 2.1-fold for both the 90- and 150-min points. For 0.75 ppm NaClO (dark histograms), and for filtration volume of 200 mL inactivation was 2.4 and 2.5 Log_{10} for the 30- and 150-min points, respectively. The inset provides a zoomed-in view of the achieved reductions over time. ClO_2 appeared to eliminate the TBC (37ºC), except for the 0.75 mg/L dose (dark histograms) where the reduction was 1.9 Log_{10}. N.d. stands for non-detectable at 100 and 200 mL of filtered sample. Bars represent the standard deviation of triplicate measurements.