Peracetic acid for secondary effluent disinfection: a comprehensive performance assessment

M. Antonelli, A. Turolla, V. Mezzanotte and C. Nurizzo

ABSTRACT

The paper is a review of previous research on secondary effluent disinfection by peracetic acid (PAA) integrated with new data about the effect of a preliminary flash-mixing step. The process was studied at bench and pilot scale to assess its performance for discharge in surface water and agricultural reuse (target microorganisms: *Escherichia coli* and faecal coliform bacteria). The purposes of the research were: (1) determining PAA decay and disinfection kinetics as a function of operating parameters, (2) evaluating PAA suitability as a disinfectant, (3) assessing long-term disinfection efficiency, (4) investigating disinfected effluent biological toxicity on some aquatic indicator organisms (*Vibrio fischeri*, *Daphnia magna* and *Selenastrum capricornutum*), (5) comparing PAA with conventional disinfectants (sodium hypochlorite, UV irradiation). PAA disinfection was capable of complying with Italian regulations on reuse (10 CFU/100 mL for *E. coli*) and was competitive with benchmarks. No regrowth phenomena were observed, as long as needed for agricultural reuse (29 h after disinfection), even at negligible concentrations of residual disinfectant. The toxic effect of PAA on the aquatic environment was due to the residual disinfectant in the water, rather than to chemical modification of the effluent.

Key words | agricultural reuse, ecotoxicity, kinetics, microbial regrowth, peracetic acid, wastewater disinfection

M. Antonelli (corresponding author)
A. Turolla
C. Nurizzo
Politecnico di Milano,
DICA – Environmental Section,
Piazza Leonardo da Vinci 32,
20133 Milano,
Italy
E-mail: *manuela.antonelli@polimi.it*

V. Mezzanotte
Università degli Studi di Milano Bicocca,
DISAT,
Piazza della Scienza 1,
20126 Milano,
Italy

INTRODUCTION

In Italy the use of chlorine-based disinfectants has been strongly constricted due to the risks related to Disinfection By-Products (DBPs) occurrence. Regulations on wastewater reuse set the limit for total trihalomethanes at 0.03 mg/L, which practically excludes chlorination as a main disinfection in Waste Water Treatment Plants (WWTPs), involving the need for alternative solutions. Among these, peracetic acid (PAA) is a broad-spectrum disinfectant, not noted for DBPs generation at low dosages (<5–10 mg/L) (Crebelli *et al.* 2005; Nurizzo *et al.* 2005). One of the main advantages of PAA is the possibility of an easy retrofit of sodium hypochlorite (NaOCl) disinfection equipment, which is almost always present in existing WWTPs, without expensive and structural interventions. This has particularly favoured the spread of PAA disinfection for WWTP upgrade in comparison with other disinfection technologies, such as UV irradiation, requiring the implementation of dedicated facilities and filtered effluents.

Even though antimicrobial activity of PAA has been thoroughly investigated and reported in many papers (*inter alia*: Kitis 2004; Koivunen & Heinonen-Tanski 2005), many process features ask for a comprehensive approach, such as PAA decay and disinfection kinetics, the potential occurrence of microbial regrowth, due to the presence of acetic acid in disinfected effluents, and direct and indirect ecotoxicological effects on the aquatic environment. These aspects are critical for determining PAA applicability, especially for agricultural reuse of treated effluents, for which very low bacterial concentrations are requested (Italian standard is 10 CFU/100 mL for *Escherichia coli* at point of use).

This study is a review of extensive research aimed at assessing the performance of PAA disinfection on secondary effluents, integrated with new data about the influence of a preliminary PAA flash-mixing step on disinfection efficiency. Experiments were carried out at bench and pilot scale using wastewater from two WWTPs located in the

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Milan urban area (Italy) starting from 2005, adopting *E. coli*, faecal coliform bacteria and total heterotrophic bacteria (not reported here) as indicators. The purposes of the research were: (1) determining PAA decay and disinfection kinetics as a function of operating parameters (active concentration, contact time, mixing conditions), (2) evaluating the suitability of PAA as a disinfectant in terms of inactivation efficiency, both for surface water discharge and agricultural reuse, (3) assessing long-term disinfection efficiency with respect to case-specific effluent reclamation, (4) investigating effluent ecotoxicity due to PAA dosage, by measuring the extent of damage to species representative of various trophic levels in the aquatic ecosystem (*Vibrio fischeri*, *Daphnia magna*, *Selenastrum capricornutum*), (5) comparing PAA with conventional disinfectants, such as NaOCl and UV irradiation.

METHODS

The secondary effluents of two municipal WWTPs were used, whose treatment schemes were based on primary treatment, pre-denitrification/nitrification and final clarification. Effluents were filtered at pilot scale (rapid sand filtration: $D_{10} = 1$ mm, filtration rate = 10.6 m/h). *E. coli* and faecal coliform bacteria were adopted as indicators.

A technical-grade PAA solution was used (Air Liquide Italia SpA; % w/w of PAA: 15, acetic acid: 17, hydrogen peroxide: 23).

Different PAA doses ($D = 1$, 2, 3, 5, 10, and 15 mg PAA/L) and six contact times ($t_C = 5$, 6, 12,18, 36, 42, and 54 min) were tested to study disinfectant decay both in tap water (dechlorinated before use) and in secondary effluents at bench scale. The same D and t_C were used for effluent disinfection tests at bench scale. Trials were performed in a completely mixed batch reactor (5 L) at room temperature (20 to 22 °C) and repeated at least 5 times for each D/t_C combination, as described by Rossi *et al.* (2007).

For microbial regrowth tests, after disinfection, samples were maintained in unmixed sterile bottles at constant room temperature (20 to 22 °C) for three reference regrowth times (t_R: 5, 24, 29 h). For the 5 h regrowth time, tests were performed to evaluate both the potential (residual PAA quenched by sodium thiosulphate 0.05 mol/L) and real (no residual PAA quenching) regrowth conditions. Each combination of $D/t_C/t_R$ was repeated four to eight times. Detailed information is reported in Antonelli *et al.* (2006).

Disinfection tests for ecotoxicological assessment using microbiotests were performed at bench scale in a completely mixed batch reactor (1 L) at room temperature (20 to 22 °C). PAA dose and contact time were fixed at 2 mg/L and 1 h. Bioassays were performed before and after disinfection, both on unquenched and quenched samples, using Microtox and commercially available kits (Ecotox LDS, Italy). Bioassay characteristics and experimental procedures are described in Antonelli *et al.* (2009).

Pilot scale tests were conducted in two in-series chicane contact tanks (Q up to 4.5 m^3/h), in which PAA doses (2 to 25 mg/L) and contact times (6 to 54 min) were tested. A series of tests was carried out dosing PAA at the inlet of the first tank without any previous mixing, while another included a flash-mixing step. The hydraulic behaviour of contact tanks had been verified by tracer tests using sodium chloride to set the operating conditions to better approach ideal plug-flow.

Sodium hypochlorite benchmark tests were conducted in the same pilot scale unit, while for UV disinfection a pilot reactor equipped with six low-pressure mercury lamps was used. NaOCl doses varied from 0.5 to 7.5 mg/L for selected contact times (6 to 54 min), while UV doses ranged from 2 to 90 mJ/cm^2. Details are reported in Mezzanotte *et al.* (2007) and Antonelli *et al.* (2008).

Influent and effluent characteristics (pH, turbidity, TSS, TOC, COD, UV absorbance at 254 nm, residual disinfectant concentration, bacterial concentration) were analysed according to *Standard Methods* (APHA 1998); for *E. coli* and faecal coliform bacteria, a plate count technique based on a membrane filtration procedure was used: faecal coliform bacteria were enumerated, as blue-green colonies, after 24 h incubation at 44 °C on the chromogenic substrate C-EC Agar (Biolife, Italy) and *E. coli* colonies were evidenced on the same plates through Wood lamp (365 nm) fluorescence.

RESULTS AND DISCUSSION

Doses and contact times were chosen with the aim of testing a wide range of conditions while remaining within acceptable cost/efficiency ratios.

PAA decay

It was verified that, unlike chlorine, the decay of pure PAA takes place over time even in tap water, leading approximately to a 25–30% decrease after 1 hour. Therefore, PAA decay occurs independently from the oxidation demand of wastewater, probably due both to hydrolysis and to the

presence of decay promoters such as metals (Fe^{2+} and Mn^{2+}, respectively 200 ± 80 μg/L and 30 ± 5 μg/L), which can easily be oxidized by PAA. A first-order kinetics seems appropriate to describe experimental data for tap water ($n = 63$, $R^2 = 0.983$, $k = 0.007$), while the best fitting equation for wastewater was obtained using first-order kinetics modified with a term of initial oxidative consumption, according to Haas & Finch (2001):

$$D_t = (D_0 - \mathrm{OC}) \cdot e^{-kt}$$

where: D_t [mg/L] = residual disinfectant concentration at time t, D_0 [mg/L] = applied disinfectant dose, OC [mg/L] = initial oxidative consumption, k [min^{-1}] = kinetic constant, t [min] = time, corresponding to t_C.

Regression statistics for secondary effluent experimental data ($n = 262$) gave $R^2 = 0.980$, $D = 0.415$ mg/L, $k = 0.007$ min^{-1}. All parameters, both for tap water and secondary effluent, were statistically significant at a 95% confidence level.

Similar kinetic constants were found for tap water and for secondary effluent, confirming that decay is not due to oxidable organics, determining only the shifting of the curve by the OC factor. Summarizing, both PAA natural decay and oxidative consumption should be considered in the choice of the disinfectant dose in designing a PAA disinfection unit.

Disinfection efficiency

The PAA disinfection displayed a log reduction from 1 to nearly 5 of *E. coli* and faecal coliform bacteria, depending on initial bacterial concentration, residual PAA and contact time.

Starting from two different initial bacterial concentrations, as shown in Figure 1, the expected log reduction of *E. coli* is reported for various D/t_C combinations. These values are calculated as an average of the log reduction values experimentally obtained, for each PAA dose and contact time, splitting the data set on the basis of *E. coli* concentration in the secondary effluent, representative of low (10×10^3 CFU/100 mL) and high (50×10^3 CFU/100 mL) initial microbial concentrations. These two values were chosen to take into account the peculiar situation in the Milan urban area, where effluents are particularly diluted. Note that it is always important to consider not only log reduction, but also final bacterial concentration corresponding to the specific disinfectant concentration and contact time; in fact, for high Ct values, the measured residual bacterial concentration tends to zero, so that log reduction is arithmetically limited by the value of the initial microbial concentration, and this leads to underestimating the real disinfection potential. This is well outlined in Figure 1 where high Ct values lead to lower log reduction when the initial *E. coli* concentration is 10×10^3 CFU/100 mL. For complying with the Italian standard for discharge in surface water (5,000 CFU/100 mL for *E. coli*), with 1 or 2 mg/L of PAA, a contact time over 18 min is necessary; at higher doses, a contact time ≥ 6 min is adequate. For complying with the more stringent standard for agricultural reuse, doses over 5 mg/L and longer contact times are required. Actually, these values are largely protective because they comply with the standards in 100% of cases, since all data above the standards have been rejected, although some of them could have been accepted considering the typical variability of microbiological analyses. Finally, the considered PAA doses and contact times are comparable to those typical of chlorine disinfection, allowing a 4-log reduction of *E. coli* with doses from 5 to 10 mg/L and contact times from 35 to 50 min. For average initial *E. coli* concentrations about 10×10^3 CFU/100 mL, the conditions required for disinfection are less severe than for higher initial concentrations.

To evaluate disinfection efficiency as a function of operating parameters, the usually adopted Ct approach is not

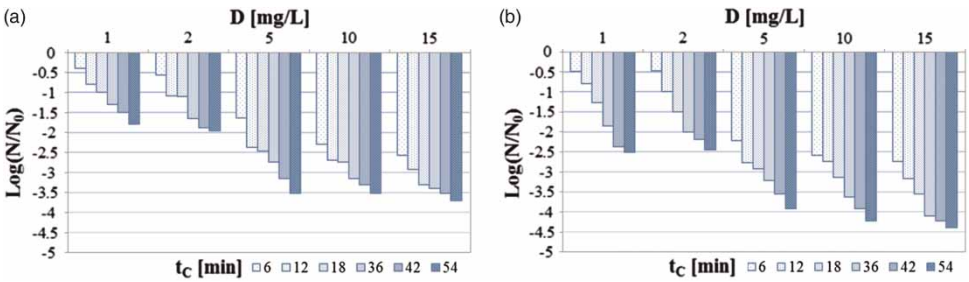

Figure 1 | Expected log reduction of *E. coli* as a function of PAA dose and contact time, starting from two different *E. coli* concentrations, respectively 10,000 CFU/100 mL (a) and 50,000 CFU/100 mL (b).

adequate, as can be noted from the high dispersion of data reported in Figure 2.

The main factor which probably affects data dispersion is the fact that the use of final residual PAA as a process parameter is not appropriate, since PAA naturally decays over the contact time, so the available disinfectant concentration varies, in opposition to what occurs with NaOCl. Consequently, different disinfection kinetic models (Table 1) were evaluated by non-linear multivariate analysis. Selleck and Chick-Watson models gave unsatisfactory results ($R^2 < 0.4$), confirming the inadequacy of some classical disinfection kinetic models in fitting experimental data when disinfectant decay is significant over process time. In fact, none of the inactivation models listed in Table 1 considers the possibility for disinfectant demand or decay, with the exception of the S-model, proposed for PAA disinfection by Profaizer (1998). Hom's model appears to be more adequate, but the active concentration to be used for fitting has to be carefully chosen, since the combination of Hom's model with the first order decay kinetics is possible, but leads only to a numerical solution. A valuable solution has been found in accounting for PAA decay by considering the PAA mean concentration during contact time, calculated as the average of PAA applied dose and final residual concentration, as the independent variable of the model. This procedure required only the knowledge of the residual PAA concentration, without needing decay kinetic parameters. Further details over data processing are reported in Rossi *et al.* (2007).

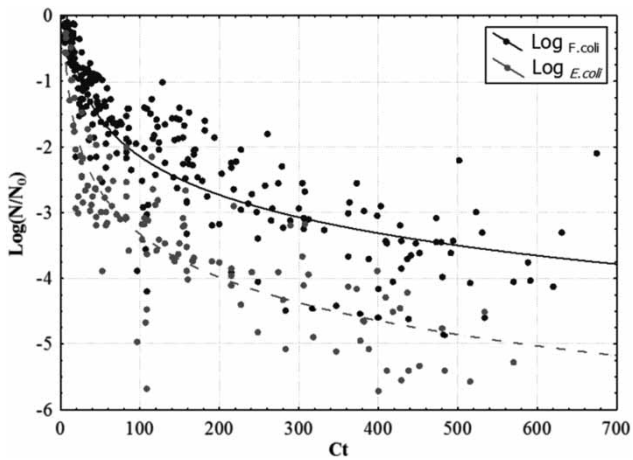

Figure 2 | Log reduction by PAA for *E. coli* and faecal coliform bacteria (*E. coli*, dashed line) as a function of Ct.

The inactivation kinetics of *E. coli* and faecal coliform bacteria can be satisfactorily modelled either by Hom's model accounting for PAA decay (Figure 3) or by the S-model: the estimated correlation coefficients for the former were 0.88 and 0.90, for *E. coli* and faecal coliform bacteria respectively, while they were 0.86 and 0.92 for the latter. A peculiar behaviour can be outlined for lower doses and contact times, especially for *E. coli*, for which an initial lag in the PAA disinfection was observed. For higher doses, inactivation increases rapidly in the early stages of the process and then follows an asymptotic trend, while, for lower doses, the inactivation trend is more time-dependent. This behaviour was probably due to an initial resistance to PAA diffusion through the cell membrane, appreciable in the early process stages at low disinfectant concentrations but negligible at higher PAA doses. This resulted in a shoulder trend, which was not taken into account by the Hom's model (but shown by data in Figure 3), but effectively described by the S-model. Actually, the S-model may better describe a complex inactivation trend, including an initial shoulder and a final tailing-off, but the correct choice of the four parameters is definitely a crucial point, since the model is less stable and robust than Hom's, especially when all the data are considered together and these data have very different behaviours, as in this case for low (1 and 2 mg/L) and high (over 5 mg/L) PAA doses.

Pilot scale tests confirmed bench scale results, giving comparable disinfection efficiency for the selected D/t_C combinations. As observed in lab tests, contact time has a strong influence on disinfection efficiency at low doses.

No significant variation in the inactivation of the selected indicators was observed over 5 mg/L dose and for contact times over 18 min as a function of the PAA dosing procedure (with and without a preliminary flash-mixing step to promote homogenous and fast distribution of PAA). For lower doses and shorter contact times

Table 1 | Wastewater disinfection models

Selleck *et al.* (1978)	Chick (1908)	Hom (1972)	S-model (Profaizer 1998)
$\frac{N}{N_0} = \left(\frac{C \cdot t}{b}\right)^{-d}$	$\log\frac{N}{N_0} = -L_s \cdot C^n \cdot t$	$\log\frac{N}{N_0} = -k \cdot C^n \cdot t^m$	$\log\frac{N}{N_0} = -\frac{k \cdot C^n}{1 + (h/(C \cdot t))^m}$

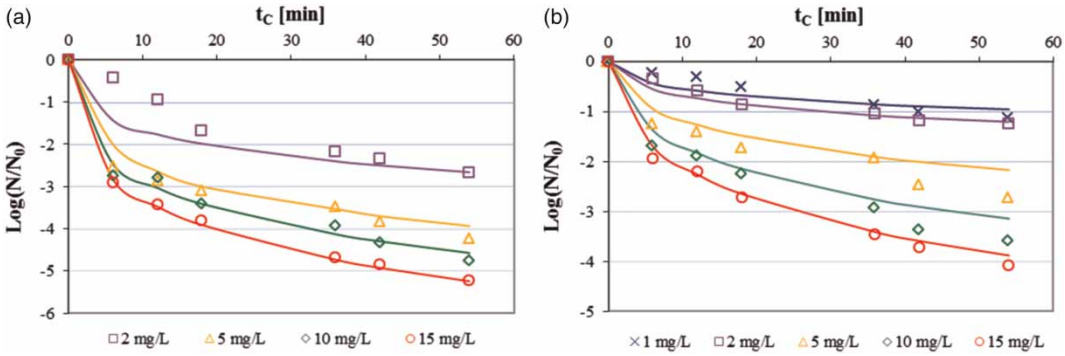

Figure 3 | Log reduction vs. time: mean of experimental data (dots) and Hom's model (unbroken lines) for *E. coli* (a) and faecal coliform bacteria (b).

pre-mixing allowed an improvement of disinfection performance, as shown in Figure 4, especially for high *E. coli* initial counts ($>20\times10^3$ CFU/100 mL). The influence of contact time was relevant for trials without pre-mixing, while in the case of pre-mixing it became relevant only for the lower doses.

Potential bacterial regrowth

The role of residual PAA in preventing bacterial regrowth was studied by comparing bacterial counts 5 h after the end of the disinfection treatment in samples with and without (quenched) residual PAA. Without any residual PAA, the log reductions observed 5 h after disinfection were comparable to those obtained immediately after disinfection for all doses and contact times, confirming the irreparability of the damages caused to coliform bacteria and consequently the PAA bactericidal efficiency, as already reported by Santoro *et al.* (2007). When residual PAA was not quenched, the disinfection process went on, with a further reduction by about 1 to 2.5-log of faecal coliform bacteria and 1 to 2-log of *E. coli*, depending on the D/t_C combinations. Regrowth was also studied over longer periods (24 and 29 h), to simulate the time needed to transfer reclaimed water to the irrigation area and for its use. For quantifying bacterial regrowth the index recommended by Kelner (DR, degree of reactivation) in 1951 was adopted:

$$\mathrm{DR} = \frac{N_r - N_d}{N_0 - N_d}$$

where: N_0 = initial number of microorganisms, N_d = number of microorganisms surviving disinfection, N_r = number of microorganisms after a fixed regrowth time.

This index represents the fraction of the initially inactivated bacteria (N_0–N_d) that are subsequently reactivated (N_r–N_d). In case of regrowth DR is positive, while it is about 0 both if $N_r = N_d$ (no regrowth) or if $N_0 >> N_d$ (a high level of disinfection is reached so that any possible further increase of N_r with respect to N_0 is not appreciable). Mean values of DR for faecal coliform bacteria are reported in Figure 5. Similar results were obtained for *E. coli*.

No significant regrowth was observed: DR values were comparable for any of the tested combinations $D/t_C/t_R$, confirming again that the overall PAA effect is long-lasting, despite the potential reparability of damage and the release

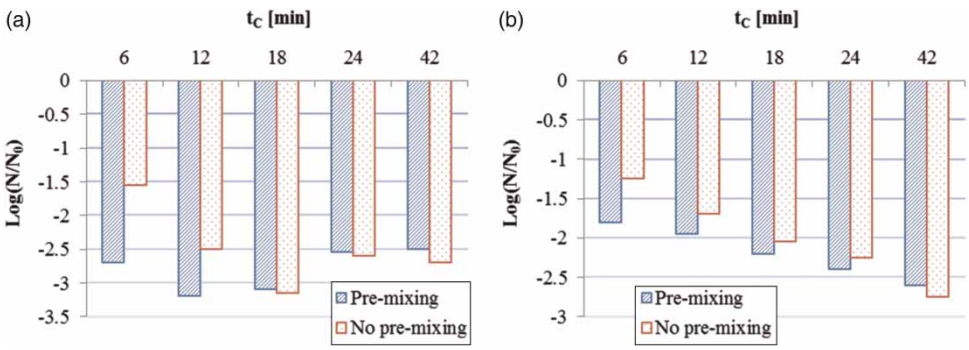

Figure 4 | Average log reduction of *E. coli* (a) and faecal coliform bacteria (b) at 5 mg/L PAA dose and different contact times, with and without pre-mixing.

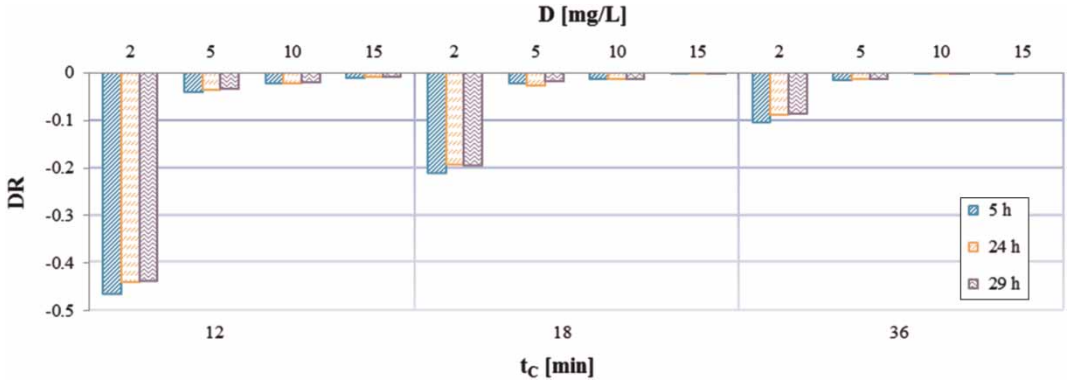

Figure 5 | Degree of reactivation (DR) for faecal coliform bacteria vs. different initial PAA doses (D), contact times (t_C) and regrowth times (in the legend).

of acetic acid. It is important to stress that: (1) PAA disappeared completely in the first 5–10 h, for the lower PAA doses, so no further disinfection occurs; (2) complete disinfection is obtained in the first 5 h for the higher PAA doses. Detailed information is reported in Antonelli *et al*. (2006).

Potential toxicity of disinfected effluents

Microbiotests were used to assess the effluent toxicity before and after disinfection. The effluent before disinfection had no toxic effect on the test organisms (Figure 6), even if for *Vibrio fischeri* the inhibition effect was negative, meaning that the bacteria were more active in the biological effluent than in the culture media (higher bioluminescence emission). This was probably due to the presence in the effluent of different potentially toxic substances, including mostly long-chain-aldehydes, which are likely to enhance the light emission, modifying the organisms' response, as discussed by Lyzen & Wegrzyn (2005). Quenching of residual PAA permitted us to assess the toxicity associated with the potential variation of the chemical composition of the disinfected samples, which could not be detected by the conventionally monitored physical–chemical parameters. Experimental results in Figure 6 showed a slight increase in the inhibition of *Vibrio fischeri* and no significant effect on *Daphnia magna* and *Selenastrum capricornutum*. The inhibition was always below the value set by Italian regulation for discharge in surface water (50% on *Daphnia magna*). The inhibition was about 100% for *Vibrio fischeri* and *Daphnia magna* when residual PAA was not quenched, in agreement with the fact that the PAA doses normally used give residues which are toxic for most test organisms. In contrast, no toxicity was observed on *Selenastrum capricornutum*, as confirmed by the presence of green algae in the disinfection basin of the WWTP where wastewater was collected and PAA was added.

Comparison with benchmarks

In pilot scale benchmark tests the maximum bacterial inactivation obtained by the three disinfectants was comparable, as reported in Figure 7. NaOCl was confirmed as the most effective among chemical disinfectants, not taking into account the environmental and sanitary risks that its use involves. PAA, NaOCl and UV treatments allowed us to

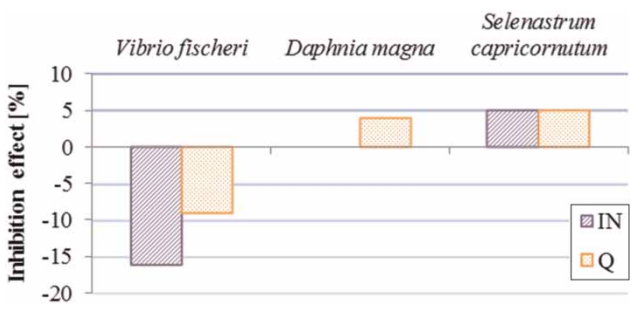

Figure 6 | Toxicity of effluents before (IN) and after disinfection (Q) by PAA (2 mg/L, 1 h, quenched PAA residual).

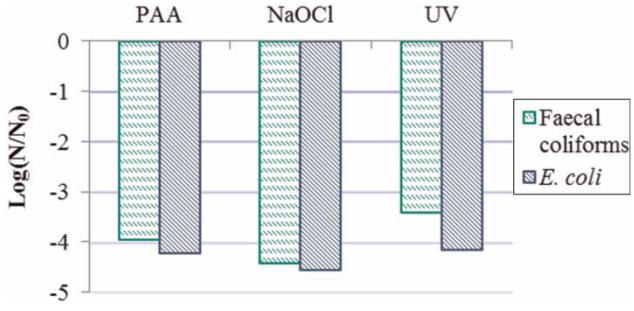

Figure 7 | Comparison between the maximum log reduction obtained by PAA (D = 15 mg/L, t_C = 36 min), NaOCl (D = 7.5 mg/L, t_C = 18 min) and UV (UV dose = 80 mJ/cm^2).

comply with Italian regulations for reuse at doses lower than the maximum tested ones. For NaOCl a 5 mg/L dose was sufficient but it generated THM concentrations exceeding the limits. UV irradiation was the best in absolute terms: in a few seconds of contact time, very low doses (10–20 mJ/cm^2) allowed complete bacterial inactivation, although bacteria regrowth after 6 h was observable for samples treated with doses below 40 mJ/cm^2 when exposed to sunlight. Further details are reported in Mezzanotte *et al.* (2007) and Antonelli *et al.* (2008).

CONCLUSIONS

Despite some disadvantages, such as natural decay and slightly less efficacy with respect to NaOCl, PAA disinfection resulted as an affordable and valuable process, able to comply with Italian regulations on wastewater discharge and reuse and competitive with NaOCl and UV irradiation. The absence of bacterial regrowth phenomena, as long as needed for agricultural reuse, and the limited toxicity effects on the aquatic environment indicated the good sanitary safety of PAA, whose bactericidal properties were demonstrated.

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