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Effect of residual H_2O_2 from advanced oxidation processes on subsequent biological water treatment: A laboratory batch study

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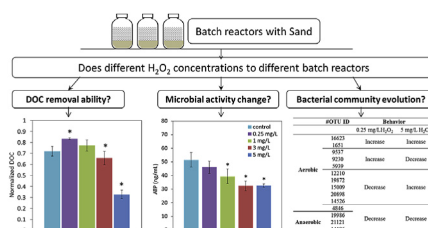
HIGHLIGHTS

- The effect of H_2O_2 on sand systems during water treatment was studied as the first time.
- DOC biodegradation was limited by 0.25 mg/L H_2O_2 and promoted by 5 mg/L H_2O_2.
- Microbial activity decreased with the increase of H_2O_2 concentrations.
- Aerobic bacteria showed different responses to H_2O_2, either sensitive or tolerant.
- Anaerobic bacteria are sensitive to H_2O_2. Their growth was limited by H_2O_2.

GRAPHICAL ABSTRACT



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ABSTRACT

H_2O_2 residuals from advanced oxidation processes (AOPs) may have critical impacts on the microbial ecology and performance of subsequent biological treatment processes, but little is known. The objective of this study was to evaluate how H_2O_2 residuals influence sand systems with an emphasis on dissolved organic carbon (DOC) removal, microbial activity change and bacterial community evolution. The results from laboratory batch studies showed that 0.25 mg/L H_2O_2 lowered DOC removal by 10% while higher H_2O_2 concentrations at 3 and 5 mg/L promoted DOC removal by 8% and 28%. A H_2O_2 dosage of 0.25 mg/L did not impact microbial activity (as measured by ATP) while high H_2O_2 dosages, 1, 3 and 5 mg/L, resulted in reduced microbial activity of 23%, 37% and 37% respectively. Therefore, DOC removal was promoted by the increase of H_2O_2 dosage while microbial activity was reduced. The pyrosequencing results illustrated that bacterial communities were dominated by *Proteobacteria*. The presence of H_2O_2 showed clear influence on the diversity and composition of bacterial communities, which became more diverse under 0.25 mg/L H_2O_2 but conversely less diverse when the dosage increased to 5 mg/L H_2O_2. Anaerobic bacteria were found to be most sensitive to H_2O_2 as their growth in batch reactors was limited by both 0.25 and 5 mg/L H_2O_2 (17–88% reduction). In conclusion, special attention should be given to effects of AOPs residuals on microbial ecology before introducing AOPs as a pre-treatment to biological (sand)

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processes. Additionally, the guideline on the maximum allowable H_2O_2 concentration should be properly evaluated.

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1. Introduction

In recent years, organic micropollutants (OMPs), such as pesticides, pharmaceutically active compounds, endocrine disrupting compounds, X-ray contrast media and personal care products, have been detected at ng/L to low µg/L concentrations in surface waters throughout the world (Kolpin et al., 2002; Stolker et al., 2004). Surface waters serve vital role to humans such as drinking water, nature, recreation and food production. These functions are susceptible to negative water quality effects from anthropogenic contaminants (Brack et al., 2017; Coppens et al., 2015). However, conventional processes and biological processes do not always provide satisfactory results for drinking water treatment (Bertelkamp et al., 2015, 2016; Paredes et al., 2016; Ruhl et al., 2014) as many organic pollutants are toxic or resistant to biological treatments. Therefore, an alternative option for such recalcitrant and biologically persistent compounds is the use of advanced oxidation processes (AOPs), widely recognized as highly efficient for water purification (Oller et al., 2011). In particular, the hydroxyl radicals (▪OH) generated by these methods have the ability to oxidise recalcitrant and non-biodegradable pollutants (Bilińska et al., 2016; Oller et al., 2011). Previous research demonstrated that the combination of AOPs, e.g. ozonation, UV/H_2O_2, ozonation/UV/H_2O_2 or photo-Fenton processes, and conventional biological processes offers an optimised treatment system to effectively remove OMPs during water treatment (Lekkerkerker-Teunissen et al., 2012; Oller et al., 2011). Integrating UV/H_2O_2 and subsequent biological activated carbon filtration may also offer a promising approach to eliminate trihalomethanes, haloacetic acids and phenol from raw surface water (Seredyńska-Sobecka et al., 2005; Toor and Mohseni, 2007). In the Netherlands, several water companies utilise intergrated AOPs with subsequent biological treatment processes. For example, Waternet in Amsterdam combines ozonation with biological activated carbon (BAC) filtration to remove OMPs during drinking water production (Bonné et al., 2002; Van Der Hoek et al., 1999). Another Dutch drinking water company, PWN, uses UV/H_2O_2 oxidation and BAC filtration to form a multi barrier approach against OMPs during drinking water production (Martijn and Kruithof, 2012). In The Hague, Dunea water utility company plans to install AOPs before managed aquifer recharge (MAR) in the dunes to form a synergistic system for the removal of OMPs (Lekkerkerker et al., 2009; Wang et al., 2016). During AOPs with O_3, H_2O_2 is present in excess to reduce the formation of the by-product bromate (Von Gunten and Oliveras, 1998; Wert et al., 2007). Therefore, H_2O_2 residuals are usually present in the effluent of AOPs.

H_2O_2 in water can function as a disinfectant with the ability to inactivate microorganisms by oxidising proteins and DNA (Apel and Hirt, 2004; Latifi et al., 2009). The growth of *A. nidulans* and *A. variabilis* was suppressed at concentrations of 0.34–3.4 mg/L H_2O_2 in dialysis culture (Samuilov et al., 1999). A study by Knol et al. (2015) suggested that H_2O_2, even in concentrations below 2 mg/L, may cause undesired effects on ecosystems in dune ponds. However, the ineffectiveness of H_2O_2 as a disinfectant, and more specifically the selective impact of H_2O_2 on microorganisms, have also been reported. For example, some phyla types had the potential to detoxify H_2O_2 in a humic lake (Glaeser et al., 2014); a concentration below 40 mg/L of H_2O_2 did not inactivate *Escherichia coli* bacteria (Labas et al., 2008); 1 mg/L H_2O_2 dosage did not decrease acetate removal by biological filters (Urfer and Huck, 1997); and H_2O_2 did not affect eukaryotic phytoplankton including green algae, chrysophytes and diatoms, even if 99% of the cyanobacterial population was reduced by H_2O_2 (Matthijs et al., 2012). Catalases are known to catalyse the conversion of H_2O_2 into water and oxygen, which is part of an adaptive response of bacteria to oxidative stress (Matthijs et al., 2012; Metz et al., 2011; Tusseau-Vuillemin et al., 2002). Some catalase-positive microorganisms, such as *Mycobacterium tuberculosis*, *Legionella pneumophila*, and *Campylobacter jejuni*, make catalase to deactivate the peroxide radicals, thus allowing them to survive (Rao et al., 2003). Another study showed additional evidence for catalase-positive bacteria that survived in the presence of H_2O_2; concentrations of H_2O_2 exceeding 0.034 mg/L were lethal for the majority of catalase-negative strains, but not for catalase-positive strains (Walczak and Swiontek Brzezinska, 2009). Additionally, even strictly anaerobic bacteria could become acclimated to normally lethal doses of H_2O_2 (Schmidt et al., 2006). Notably, the assimilable organic carbon removal efficiency slightly increased in a biological filter receiving water with 1 mg/L H_2O_2 (Urfer and Huck, 1997). Several reports on the use of H_2O_2 injection to supply oxygen into subsurface biologically active zones indicated various degrees of success when applied to contaminated aquifer remediation, but the bacterial damage by H_2O_2 has never been reported (Aggarwal et al., 1991; Tusseau-Vuillemin et al., 2002; Zappi et al., 2000), indicating the damage may be negligible. Therefore, although H_2O_2 is generally used to inactivate microorganisms in aqueous systems, some microorganisms may be able to tolerate H_2O_2 in varying concentrations and situations. In particular, the effect of H_2O_2 as a residual of AOPs on microbial activity in subsequent biological water treatment processes, such as BAC filtration and sand filtration, is not yet well understood.

Further investigation into the effects of H_2O_2 on microbial activity in sand systems is important, scientifically for microbial ecology and practically for surface water purification systems that utilise a combination of AOPs and sand systems, e.g. sand filtration or MAR in a sandy soil. The objective of this study was to evaluate in batch experiments how different concentrations of residual H_2O_2 influence sand systems with an emphasis on dissolved organic carbon (DOC) removal, microbial activity change and bacterial community evolution.

2. Materials and methods

2.1. Experimental set-up

Batch reactors with sand and water have been widely used to assess substances degradations, impact factors or influences on microbial communities (Abel et al., 2013; Lekkerkerker, 2012; Maeng, 2010; Maeng et al., 2012; Wang et al., 2016). In the present study, batch reactors (1 L glass bottles) filled with 200 g sand (grain size 0.8–1.25 mm) and 800 mL water were used to investigate the influence of H_2O_2 on microbial activity in sand systems.

Sand used in this study was collected from the top 0.5–2.0 cm of a slow sand filter used by the water utility Dunea. The top 0.5–2.0 cm (schmutzdecke) of a slow sand filter has diverse microbial communities and greatly contributes to the removal of organic matter by biodegradation processes, so this layer is considered to represent the

microbial activity of sand filtration systems (Chekol, 2009; Dizer et al., 2004).

The water used in batch reactors was prepared with demineralised water and chemical additives (33 mg Na_2HPO_4/L, 7.5 mg NaH_2PO_4/L, 22 mg K_2HPO_4/L, 140 mg $CaCl_2$/L, 0.031 mg $FeCl_3$/L, 0.032 mg NH_4Cl/L, 40.75 mg $MgSO_4$/L, 17.823 mg $NaNO_3$/L, 0.00114 mg $MnCl_2$/L, 82 mg CH_3COONa/L) and simulated the pre-treated surface water (after AOPs) of Dunea as used in drinking water production. Additionally, in order to have residual DOC and avoid bacterial starvation conditions, the carbon source (as sodium acetate) in the batch reactors was 22 mg/L DOC which was around 5 times higher than that found in pre-treated surface waters. However, in practice, the pre-treatment by AOPs will increase the amount of biodegradable organic matter and may lead to increased microbial activity in the influent water of the subsequent biological process, probably two to three times higher than biological treatment systems without the pre-treatment AOPs (Pharand et al., 2014). Table 1 shows the composition of water in batch reactors. The H_2O_2 solution was prepared from a 30% standard solution (Merck, Germany). All the solutions used in this study were prepared using water from a Millipore Milli-Q system. All chemicals were of analytical grade purity (AR grade $\geq$ 99% purity or better).

2.2. Experimental processes

The experimental processes are presented in Fig. 1. 18 batch reactors with 200 g sand and 800 mL water were used. The adaptation of microbial communities found on the sand to laboratory conditions was achieved by refreshing water every 5–7 days until steady state conditions were reached with respect to DOC removal calculated as $DOC_{ending}/DOC_{initial}$ (Lekkerkerker-Teunissen et al., 2012; Maeng, 2010). $DOC_{initial}$ was measured at the beginning just after refreshing water and DOC_{ending} was the DOC concentration in the batch reactor just before refreshing water. Fig. S1 (supplemental information 1) shows the results for normalised DOC removal during the ripening period. DOC data show that steady state conditions were achieved after around two months.

After ripening the reactors, H_2O_2 spiking experiments started. The research of Lekkerkerker (2012) and Knol (2012) showed that a 6 mg/L H_2O_2 dosage was adequate to form sufficient •OH for oxidation in AOPs so that the residual H_2O_2 concentration in effluent water of AOPs will not exceed 6 mg/L. Therefore, different dosages of H_2O_2 were added to reactors to result in final concentrations of 0.25, 1, 3, 5 mg/L in 15 non-autoclaved batch reactors after water refreshing. To distinguish DOC oxidised by H_2O_2 directly from DOC biodegradation, 3 additional reference batch reactors were autoclaved at 121 °C for 40 min to inactivate microbes and then dosed with 5 mg/L H_2O_2.

To avoid heavy damage to microbial communities from a high H_2O_2 load and also to facilitate the gradual adaptation of the microorganisms to the spiked H_2O_2, H_2O_2 was dosed into the 15 non-autoclaved batch reactors once per day during the initial shock load phase (phase 1, 6 days), 3 times per day during the intermediate phase (phase 2, 6 days), and finally as a continuous load using a pump (phase 3, 6 days). For phase 3, H_2O_2 concentrations of 0.25 mg/L, 1 mg/L, 3 mg/L and 5 mg/L groups were realised in the reactors by pumping 9 mL of feed solutions of 133.4, 530, 1590 and 2650 mg/L into these reactors respectively. DOC in each batch reactor was returned to 22 mg/L every 5–7 days by refreshing the reactor with water containing sodium acetate during the ripening phase, while during the H_2O_2 spiking period (phase 1, 2 and 3) the same DOC concentration, 22 mg/L, was reached every 2 days by dosing appropriate amounts of sodium acetate to each batch reactor to avoid the impact of DOC concentration differences among batch reactors on microbial community structure. Considering the accumulation of bacterial metabolites with time, the water in the batch reactors was refreshed at the end of each phase. 15 mL water samples for DOC analysis were collected 9–11 times to investigate the potentially different DOC removal responses to H_2O_2 over time. To estimate the H_2O_2 decomposition, 8 mL H_2O_2 water samples were collected on the first day after H_2O_2 was added. Adenosine triphosphate (ATP) samples were collected from the water instead of the sand to prevent disturbance and heavy loss of sand in our reactors. A previous study, described in detail in supplemental information 2, showed a positive correlation between ATP in the water and in the sand (Fig. S2 in supplemental information), so ATP in the water can be positively correlated with ATP in the sand. 1 mL water samples for adenosine triphosphate (ATP) analysis were taken 4–10 times in each phase to assess the microbial population responses to H_2O_2 over time. At the beginning of the spiking experiment, both DOC and ATP sampling frequencies were high in order to determine the optimal sampling time. To investigate the effect of low (0.25 mg/L) and high (5 mg/L) H_2O_2 concentrations on microbial composition and diversity in sand systems, sand samples were taken from the control (0 mg/L H_2O_2), 0.25 mg/L and 5 mg/L groups at the end of the experiment for 16-S pyrosequencing measurement (Huang and Chen, 2004).

To distinguish DOC abiotic removal by directly oxidation by H_2O_2 from biotic removal in sand systems, 5 mg/L H_2O_2 was dosed to 3 autoclaved batch reactors as references at the beginning. DOC and H_2O_2 concentrations were measured at 5 different time points (T = 0 h, 8 h, 24 h, 48 h, 72 h). ATP was measured at t = 0 h, 24 h, 48 h and 72 h to confirm the elimination of biological activity in the autoclaved batch reactors. ATP was present in the autoclaved batch reactors in the range of 0.04–0.06 ng/mL during the 72 h testing period, which indicated bacterial inactivation. The experiment was finished in 3 days in order to minimize growth of bacteria from the surrounding environment inside the batch reactors, which were in contact with air. DOC and H_2O_2 results in autoclaved batch reactors within 3 days were sufficient to distinguish DOC abiotic removal from biotic removal.

All batch reactors were placed in a dark, temperature (12 ± 0.5 °C) controlled room and left uncovered so that the air could enter the batch reactors. All batch reactors were prepared and sampled in triplicate.

2.3. Analysis

2.3.1. DOC

DOC was measured with a Shimadzu TOC-VCPH/CPN analyser with a standard deviation of 0.1 mg/L immediately or within one day after sampling. First, all samples were diluted one time using deionised water, then 30 mL of the diluted mixture was measured at constant temperature (20 °C) after being filtered through 0.45 μm filters (SPARTAN™, Whatman, Germany) that had been flushed twice with deionised water. To remove the inorganic

Table 1
The composition of water in batch reactors.

O_2 (mg/L)	pH	NH_4^+–N (mg/L)	NO_3^-–N (mg/L)	SO_4^{2-} (mg/L)	Fe^{3+} (mg/L)	Mn^{2+} (mg/L)	DOC (mg/L)
9 ± 1.0	7.8 ± 0.3	0.00847	2.9 ± 0.1	30.6 ± 2	0.0106	0.0005	22

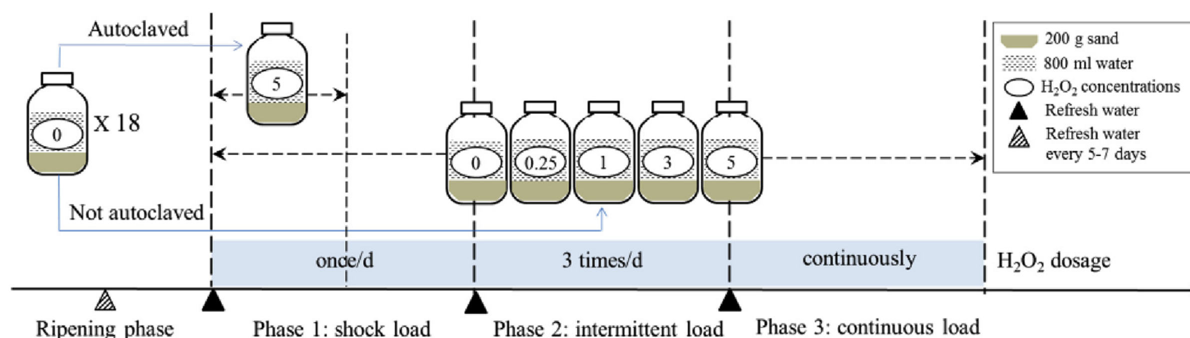

Fig. 1. Batch reactors with different operation conditions (n = 3). The ripening phase lasted for 2 months, then three batch reactors were autoclaved while the other fifteen batch reactors were not autoclaved. 5 mg/L H_2O_2 was dosed to the autoclaved reactors, and different concentrations of H_2O_2 (0, 0.25, 1, 3, 5 mg/L) were dosed to non-autoclaved reactors. Each H_2O_2 dosage phase was 6 days long.

carbon, samples were acidified by adding 1.6 mL 2 mol/L HCl (Sigma-Aldrich) before measurement.

2.3.2. ATP analysis

ATP is used in all cells as a carrier of free energy and phosphate groups to drive many chemical reactions. It plays a key role in metabolic processes in the cells and can therefore be used as an indicator for microbial activity (Liu et al., 2013, 2016). In this study, ATP was measured as total ATP in the supernatant (Liu et al., 2013) using Quench Gone Wastewater (QG21W) test kits (Canada) and a LB9509 luminometer (Aqua Tools, France) with a standard deviation of <5%. Based on the test kit instructions, a 1 mL water sample was directly dosed into a QG21W extraction tube with 2 mL UltraLyse 30[21] to lyse the bacteria and release ATP. Secondly, the extraction tube and QG21 dilution tube were mixed to dilute it. Next, the luminescence reaction of sample ATP with Luminase was measured as a Relative Luminescence Unit (RLU), and finally the RLU value was compared to that of a check standard (LuminUltra's UltraCheck) and converted to ATP concentration in ng/mL.

2.3.3. H_2O_2

Hydrogen peroxide test kits (1.18789.0001, VWR company) with a detection range of 0.015–6.00 mg/L were used for water-phase H_2O_2 measurements because of ease of operation, the rapid decomposition of H_2O_2 and accuracy of results. Since the sand water mixture in this experiment was turbid, 8 mL was pipetted into the reaction cells after filtration through 0.45 μm filters. After 10 min, the sample was transferred to a 10/20 mm rectangular cell and measured in a photometer (Spectroquant NOVA 60).

2.3.4. Bacterial qualitative analysis-pyrosequencing

At the end of experiments, 5 g sand was sampled from selected groups (0 mg/l, 0.25 mg/l, 5 mg/l) and bottles (duplicates). DNA was extracted using a Power Soil kit according to the manufacturer's instructions, and the 16S rRNA profiling was performed by 454 pyrosequencing (Medisch Moleculair Microbioloog Streeklab, the Netherlands). The primers used were GACACTATAGGATTAGA-TACCCBRGTAGTC (forward) and CACTATAGGGTCACGRCACGAGCT-GACGAC (reverse). Around 3000 readers were obtained. Obtained sequences were trimmed, merged alignments of the sequences were aligned via the infernal aligner from the Ribosomal Database Project (RDP) pyrosequencing pipeline, and the NAST alignment tool from Greengenes was obtained via the software. The RDP Classifier was used for the taxonomical assignments of the aligned 454 pyrosequencing at the 97% confidence level. The bacterial communities from all samples were analysed for the number of operational taxonomic units (OTUs), species richness and biodiversity using the QIIME program.

2.3.5. Statistical analysis

Significant difference in individual parameters between water and H_2O_2 treatments (n = 6) was analysed with one-way ANOVA tests using SPSS 17.0 (SPSS, Chicago, IL, USA). A difference was considered statistically significant at $p < 0.05$. As described in section 2.2, to maintain the same DOC concentration in all batch reactors, DOC was recovered to 22 mg/L by dosing different amounts of the carbon source every 2 days, so cumulative DOC in batch reactors was different and may therefore lead to different total DOC removals. The partial correlation analysis between DOC concentrations and DOC accumulations and H_2O_2 dosages was applied to explore if DOC removal differences between each H_2O_2 dosage groups were caused by different H_2O_2 dosages or different carbon source accumulation.

2.3.6. Other analyses

Dissolved oxygen, pH and temperature were measured with a multimeter (Sentix 41 probe, Multi 340i, WTW, Germany).

3. Results

3.1. DOC removal and H_2O_2 decomposition

To show the effect of DOC calibration every two days in each phase and refreshing the reactor water at the end of each phase, DOC fluctuations of the control group and 5 mg/L H_2O_2 group are presented as an example in Fig. 2-a. To illustrate the influence of H_2O_2 on DOC removal in greater detail, Fig. 2b, c and d present the DOC removal of each H_2O_2 dosage group.

Two phenomena can be observed in Fig. 2-a. Firstly, normalised DOC as DOC_t/DOC_0 (initial DOC concentration) in the control group decreased to 21–35% at the beginning (the first 2 days) of each phase, 58–73% in the middle (the second 2 days) and the end (the last 2 days) of each phase. Every 5–7 days, the reactor water was refreshed and DOC_0 was returned to 22 mg/L in each batch reactor to ensure sufficient growth space and nutrients. DOC removal between the control and 5 mg/L groups had no apparent difference during phase 1 (H_2O_2 shock load), while DOC removal in the control group became slightly lower than 5 mg/L group during phase 2 (H_2O_2 intermittent load). This phenomenon became more apparent in phase 3 (H_2O_2 continuous load). The same pattern was observed for the other H_2O_2 dosage groups: no obvious difference of DOC removal, 29%–33%, between the H_2O_2 dosage groups was observed at the end of phase 1 (Fig. 2-b); interestingly, DOC removal slightly increased with the increase of H_2O_2 dosage at the end of phase 2

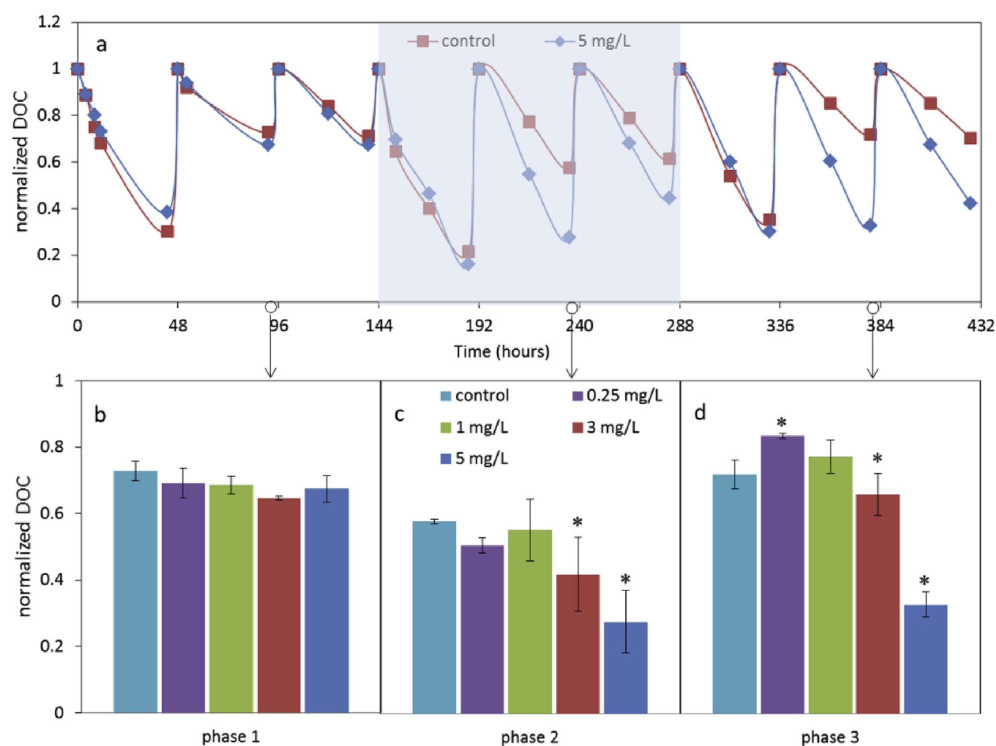

Fig. 2. Normalised DOC concentrations in batch reactors (n = 3) over time (a), at the middle of phase 1 with shock load (b), phase 2 with intermittent load (c) and phase 3 with continuous load (d). The light blue shadow highlights phase 2. $p > 0.05$ for Fig. 2-b, $p < 0.05$ for Fig. 2-c, and $p < 0.05$ for Fig. 2-d. * signifies a significant difference from the control ($p < 0.05$). (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

(Fig. 2-c), and this trend became even more apparent at the end of phase 3 (Fig. 2-d).

To assure that the above DOC removal differences between each H_2O_2 dosage groups were indeed caused by different H_2O_2 dosages and not by the cumulative differentiation in DOC dosage between the groups, Table S1 in supplemental information 3 presents partial correlations between the normalised DOC concentration and cumulative DOC dosage and H_2O_2 dosage. These correlations clearly indicate that the manner of dosing DOC – returning to 22 mg/L every two days – did not interfere with the objective of the experiment.

Based on the result of variance analysis, 0.25 mg/L H_2O_2 significantly limited DOC removal by 11% while 3 and 5 mg/L H_2O_2 promoted DOC removal by 6% and 33% respectively in comparison with the control group (Fig. 2). The results above suggest that the DOC removal in batch reactors was enhanced under the presence of H_2O_2 after an adaptive period of several days.

In non-autoclaved batch reactors, the H_2O_2 decomposition in different H_2O_2 dosage groups is presented in Fig. 3-a. H_2O_2 initial concentrations in the range of 0.25–1 mg/L decomposed to below the detection limit of 0.015 mg/L, and 3–5 mg/L H_2O_2 decomposed to 0.08 mg/L in 4 h. In the autoclaved batch reactors, however, DOC removal over time was not observed, while H_2O_2 decreased slowly from 5.4 mg/L to 2.4 mg/L within 3 days after dosing H_2O_2 (Fig. 3-b). These results illustrate that in this study DOC removal only occurred in non-autoclaved batch reactors and H_2O_2 decomposition was strongly accelerated in these reactors.

3.2. Microbial activity

ATP concentrations in the supernatant of batch reactors over the three phases are shown in Fig. 4. It can be observed that ATP concentrations in each H_2O_2 group were comparable ($p > 0.05$) during phase 1 (Fig. 4-b) and phase 2 (Fig. 4-c), while ATP in the 5 mg/L H_2O_2 group became lower than observed in the control group during phase 3 (Fig. 4-d), which may be due to the continuous H_2O_2 dosing. In phase 3 (Fig. 4-d) after the bacterial adaptive period, it appears that ATP values in high H_2O_2 concentration groups (1, 3 and 5 mg/L H_2O_2) were significantly lower than the control group (by 23%, 37% and 37%) ($p < 0.05$), and the ATP value in low concentration group of 0.25 mg/L had no notable difference compared to the control group. In phase 3, ATP decreased with the increase of H_2O_2 dosage, which indicates that a low concentration of H_2O_2 may not impact microbial activity and that only a high concentration of H_2O_2 negatively affects the microbial activity.

3.3. Microbial structure and composition

Microbial community analysis was conducted on representative sand samples from the control (0 mg/L H_2O_2), low concentration (0.25 mg/L H_2O_2) and high concentration (5 mg/L H_2O_2) groups at the end of this study (after phase 3). A broad microbial community was detected in all samples. Fig. 5 shows the phylum level bacterial community composition and their relative abundances. The bacterial communities in all groups were dominated by *Proteobacteria*, more specifically, *Betaproteobacteria* (40%–46%), and around 40% of sequences could not be assigned to any of the known phyla. The results also show that all the percentages of *Alphaproteobacteria* (from 1.45% to 2.94%), *Betaproteobacteria* (from 36.18% to 38.74%) and *Gammaproteobacteria* (from 1.75% to 3.2%) increased with the addition of 5 mg/L H_2O_2, but they did not appear to change with the addition of 0.25 mg/L H_2O_2, indicating *Proteobacteria* may have a strong resistance to H_2O_2. The abundance of *Firmicutes* became lower, from 8.84% via 8.02%–4.80%, by dosing 0.25 and 5 mg/L H_2O_2, indicating that *Firmicutes* may have low resistance to H_2O_2. At genera level, 450, 1200, and 870 genera were detected in the

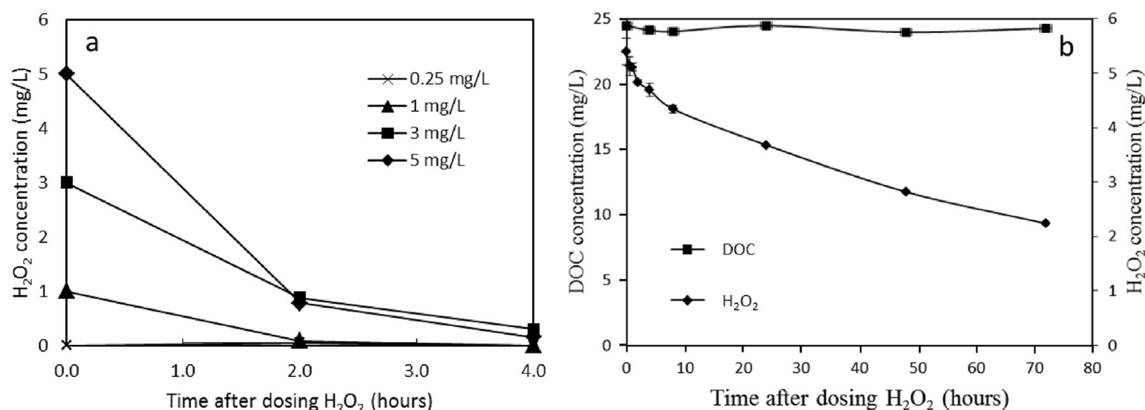

Fig. 3. (a) H_2O_2 concentrations in non-autoclaved batch reactors in the first day of the experiment (n = 3) and (b) DOC and H_2O_2 concentrations (n = 3) over 3 days after dosing 5 mg/L H_2O_2 in autoclaved batch reactors.

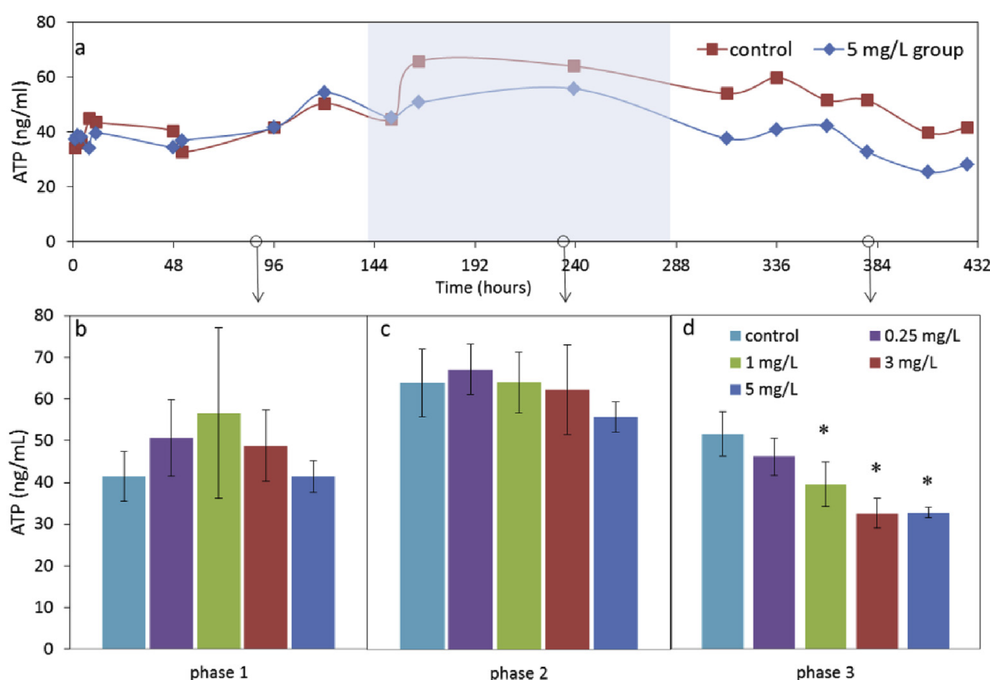

Fig. 4. ATP concentrations in the supernatant of batch reactors (n = 3) over time (a), at phase 1 with shock load (b), phase 2 with intermittent load (c) and phase 3 with continuous load (d). $p > 0.05$ for Fig. 4-b and Fig. 4-c, and $p < 0.05$ for Fig. 4-d. * signifies for significant difference from the control ($p < 0.05$).

control, 0.25 mg/L, and 5 mg/L groups, respectively.

The abundant genera (>1%) classified into four clusters are present in Table 2. It can be observed that there were not only aerobic bacteria but also anaerobic bacteria in the control group, suggesting that oxygen may have been a limiting factor for aerobic bacteria growth in batch reactors even though all batch reactors were exposed to the atmosphere. Compared with the control group, *Zoogloea* spp. (OTU 16623) and some unknown bacteria (OTU 1651) in cluster 1 increased under the presence of H_2O_2, suggesting that these bacteria have a strong tolerance to H_2O_2. 0.25 mg/L H_2O_2 increased *Zoogloea* spp. (OTU 9537) and *Comamonadaceae* spp. (OTU 9230 and OTU 5939) of cluster 2, but 5 mg/L H_2O_2 decreased their percentages, indicating that they may have a weak tolerance. For cluster 3, *Zoogloea* spp. (OTU 12210, 1987 and 15009) *and Comamonadaceae* spp. (OTU 20898 and 14526) decreased in the 0.25 mg/L H_2O_2 group while they increased in the 5 mg/L H_2O_2 group. Finally, in cluster 4, percentages of *Rhodocyclaceae* spp. (OTU 4846), *Fusibacter* spp. (OTU 19986 and 21121) *and Geobacter* spp. (OUT 14196) decreased under the presence of 0.25 mg/L H_2O_2 and further decreased under the presence of 5 mg/L H_2O_2 in comparison with the control group, suggesting sensitivity to H_2O_2. Overall, it can be seen that aerobic bacteria showed different responses to H_2O_2, either sensitive or tolerant. However, anaerobic bacteria were sensitive to H_2O_2 and their growth was limited by both 0.25 and 5 mg/L H_2O_2 (17–88% reduction).

3.4. Microbial diversity

3.4.1. Alpha diversity

Selected alpha diversity parameters (Shannon Index, Observed OTUs and Chao1) are presented in Table 3. The results indicate that a low dosage of H_2O_2 resulted in a more diverse bacterial community, whereas the high concentration dosage of H_2O_2 supressed the diversity of bacterial community.

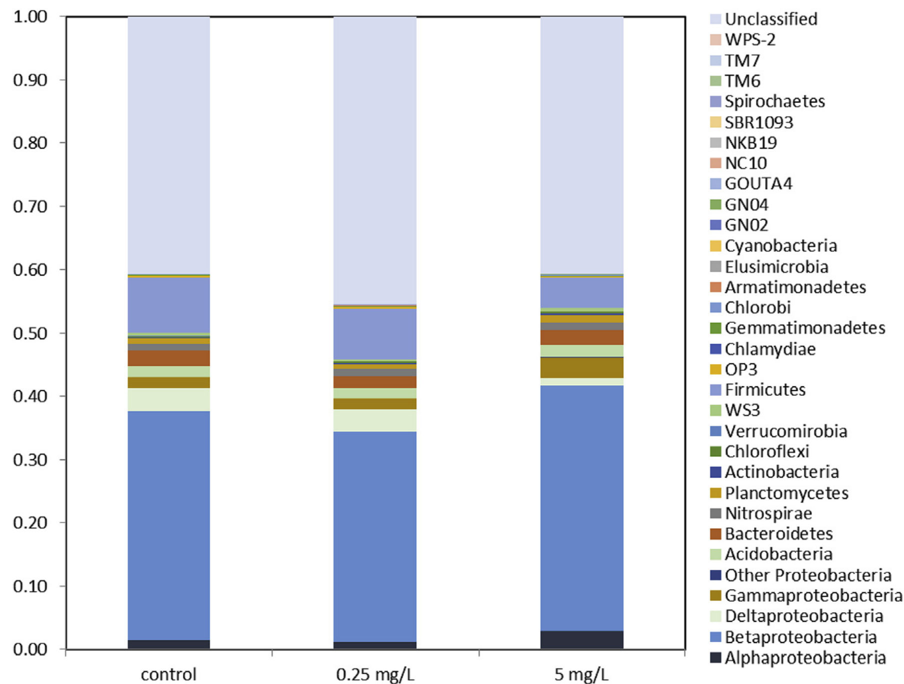

Fig. 5. The relative abundance of different phyla and subclasses in *Proteobacteria* with and without the addition of H_2O_2. The phylum of *Proteobacteria* is shown in subclasses of *Alphaproteobacteria*, *Betaproteobacteria*, *Deltaproteobacteria*, and *Gamaproteobacteria*. Samples were analysed in duplicate.

Table 2
The genera identified in the control, low H_2O_2 concentration (0.25 mg/L) and high H_2O_2 concentration (5 mg/L) groups that accounted for >1%.

		Family	Genus	#OTU ID	Control (0 mg/L)	0.25 mg/L	5 mg/L
Aerobic	Cluster 1	Rhodocyclaceae	Zoogloea	denovo16623	1.09	1.93	1.32
		Unassigned	unknown	denovo1651	0.48	1.11	0.62
	Cluster 2	Rhodocyclaceae	Zoogloea	denovo9537	1.04	1.07	0.74
		Comamonadaceae	unknown	denovo9230	1.15	1.47	0.60
		Comamonadaceae	unknown	denovo5939	1.34	1.93	0.62
	Cluster 3	Rhodocyclaceae	Zoogloea	denovo12210	6.24	2.59	6.71
		Rhodocyclaceae	Zoogloea	denovo19872	5.43	2.62	5.21
		Rhodocyclaceae	unknown	denovo15009	0.32	0.21	0.69
		Comamonadaceae	unknown	denovo20898	1.25	0.69	2.47
		Comamonadaceae	unknown	denovo14526	1.09	0.54	1.71
Anaerobic	Cluster 4	Rhodocyclaceae	unknown	denovo4846	1.08	0.55	0.52
		Acidaminobacteraceae	Fusibacter	denovo19986	4.51	3.72	2.75
		Acidaminobacteraceae	Fusibacter	denovo21121	3.50	2.90	1.61
		Geobacteraceae	Geobacter	denovo14196	1.46	1.01	0.17

The changes of their abundances as response to the addition of H_2O_2.
Cluster 1 increased at both low and high H_2O_2 dosage.
Cluster 2 increased at low H_2O_2 dosage but decreased at high H_2O_2 dosage.
Cluster 3 decreased at low H_2O_2 dosage but increased at high H_2O_2 dosage.
Cluster 4 decreased at both low and high H_2O_2 dosage.

3.4.2. Beta diversity

The comparison of the similarity of the bacterial communities was performed by principle coordinates analysis (PCoA) (Fig. 6). Results showed that bacterial communities with the same dosage of H_2O_2 clustered together while different doses resulted in different clusters, suggesting that the addition of H_2O_2 influenced the bacterial community. These changes of bacterial community may explain the different DOC removal efficiency observed based on the DOC results.

Table 3
Alpha bacterial diversity in the control, low H_2O_2 concentration (0.25 mg/L) and high H_2O_2 concentration (5 mg/L) groups.

H_2O_2 dosage (mg/L)	Shannon Index	Observed OTUs	Chao1
0 (control)	8.8 (±0.1)	909 (±10)	5700 (±300)
0.25	9.3 (±0.2)	975 (±19)	6700 (±200)
5	8.6 (±0.2)	873 (±2)	4500 (±10)

4. Discussion

4.1. Increase of DOC biodegradation under H_2O_2 presence

Since H_2O_2 is thought to disturb natural ecology by inactivating microbes and damaging flora and fauna (Knol, 2012; Kruithof et al., 2007), it is important to quench H_2O_2 residuals contained in AOPs effluent water before discharging into subsequent biological systems. This study showed that in the presence of 3 and 5 mg/L H_2O_2, the microbial activity in the water phase measured as ATP indeed decreased (Fig. 4-d), indicating that microbial activity in the sand

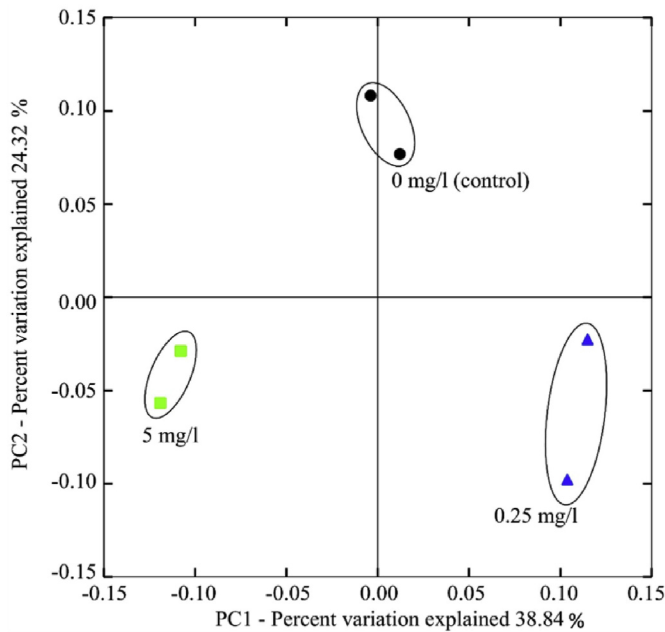

Fig. 6. Principle coordinates analysis of bacterial community similarity among different groups of samples. The control group, 0.25 mg/L group and 5 mg/L group are shown in black circles, blue triangles and green squares, respectively. Samples were analysed in duplicate.

also decreased due to the positive correlation as described in section 2.2. However, at the same time DOC removal notably increased instead of decreased (Fig. 2-d). A similar phenomenon was also observed by Urfer and Huck (1997), in which acetate removal in a biological filter receiving water with 1 mg/L H_2O_2 was slightly higher than in the control column after an adaption period of 28 days. Unfortunately, this phenomenon did not attract enough attention, and an explanation was not provided.

Although H_2O_2 may have reacted with DOC, the possibility that H_2O_2 removed DOC in this study can be excluded due to the stable DOC concentration in the autoclaved batch reactors (Fig. 3-b). Therefore, DOC removal caused by a high H_2O_2 dosage must be related to biological processes. In real sand filtration systems, it is possible that H_2O_2 oxidises organic matter into smaller molecules that can be more easily biodegraded (Chelme-Ayala et al., 2011; Metz et al., 2011), but acetate was the only carbon source in this study, and thus this reaction is not relevant. The slow decomposition of H_2O_2 in the autoclaved batch reactors can be explained by its reaction with inorganic substances attached to the sand instead of a reaction with DOC (Wang et al., 2016).

During aerobic degradation, free molecular oxygen accepts electrons released by an electron donor (e.g. soil organic carbon), which is reduced to a lower oxidation state (Morgan and Watkinson, 1992). Oxygen, potentially not present in adequate concentrations in the control group as previously described, limited the ability of aerobic microorganisms to actively degrade DOC. Fig. 3-a shows that H_2O_2 in all groups decomposed within 4 h, indicating oxygen, the decomposition product of H_2O_2, was formed quickly, and more oxygen was released in high H_2O_2 dosage groups than in low H_2O_2 dosage groups. The low H_2O_2 dosage group (0.25 mg/L) inhibited DOC biodegradation while high H_2O_2 dosage groups (3 mg/L and 5 mg/L) promoted DOC biodegradation (Fig. 2-d). It can be hypothesised that the low concentration of H_2O_2 released limited oxygen that was not sufficient to promote aerobic bacterial activity. However, high concentrations of H_2O_2 released more oxygen which served as the electron acceptor for DOC biodegradation and therefore promoted aerobic degradation. Alternatively, the increased DOC removal with H_2O_2 dosage increase could also be caused by the change in bacterial community composition, which will be discussed in section 4.2.

4.2. Effects of H_2O_2 residuals on sand bacterial community

In this study, the obtained bacterial community results confirmed that H_2O_2 residuals affected sand bacterial community composition and its alpha and beta diversity. The results confirm that the sand bacterial community is sensitive to its surrounding environments, especially to the presence of H_2O_2, which can function both as a disinfectant to oxidise proteins and DNA (Apel and Hirt, 2004; Latifi et al., 2009) and as an oxygen source to enhance aerobic bacterial growth (Hinchee et al., 1991; Tusseau-Vuillemin et al., 2002; Zappi et al., 2000). In response, the bacterial community became more diverse after adding 0.25 mg/L H_2O_2, whereas the diversity decreased when the H_2O_2 dosage increased to 5 mg/L (Table 3). Potential explanations are: 1) H_2O_2 can be detoxified by cellular enzymes (e.g. catalases and peroxidases) (Pardieck et al., 1992) and 2) oxygen from the low concentration of H_2O_2 promotes aerobic bacterial growth, although more cells are inactivated when the H_2O_2 exceeds the cellular detoxification capacity.

The different responses and resistances of OTUs to H_2O_2 dosage (genus results, Table 2) could be a complex result of H_2O_2 damage on bacterial cells (Glaeser et al., 2014), the growth promotion of oxygen from H_2O_2 decomposition (Aggarwal et al., 1991; Tusseau-Vuillemin et al., 2002) and bacterial catalase-positive property (Pardieck et al., 1992). As stated previously, cluster 1, *Zoogloea* spp. (OTU 16623) and an unknown bacteria spp. (OTU 1651), has a strong tolerance to H_2O_2, which may be explained by their catalase-positive property. Catalase is responsible for the protection, interception and repair of microorganisms against H_2O_2/■OH damage (Pardieck et al., 1992). To the authors' knowledge, the catalase-positive property of those bacteria has not been reported. However, results without a bacterial cellular catalase in this study cannot test this hypothesis, so further study is necessary. Bacteria in cluster 2 (Table 2) may have a low tolerance to H_2O_2, while the damage of H_2O_2 on bacterial cells may become a leading role with the increase of H_2O_2 concentrations up to 5 mg/L. The change of bacterial percentages in cluster 3 (Table 2) may be explained by the damage of H_2O_2 on bacterial cells playing a leading role under the presence of 0.25 mg/L H_2O_2 while the growth promotion of oxygen from H_2O_2 decomposition became larger/the same level than the control group. A notably large reduction of the bacterial percentage occurred in cluster 4 (Table 2), therefore, those bacteria may be catalase-negative. *Fusibacter* and *Geobacter* are anaerobic bacteria that have been found in anaerobic conditions in soils and aquatic sediment (Lovley et al., 1987). Notably, percentages of all anaerobic bacteria, *Fusibacter* spp. (OTU 19986 and 21121) and *Geobacter* spp. (OTU 14196) were largely lowered under the presence of low and high concentrations H_2O_2, which can be explained by oxygen released by H_2O_2, inhibiting their growth and/or H_2O_2, damaging bacterial cells and DNA.

The observed changes in bacterial community caused by H_2O_2 residuals may influence the organic matter removal in sand systems since microbial degradation and assimilation play a dominant role in the attenuation of organic compounds (Amy and Drewes, 2007). This can be confirmed by the above DOC removal efficiencies of different groups: the highest DOC removal was found in the 5 mg/L H_2O_2 group, while the lowest removal was found in the 0.25 mg/L H_2O_2 group. It is hard to conclude which genus or species contributed to DOC removal change in low and high H_2O_2 dosage groups, but the following hypothesis is provided. Bacteria of cluster

3 had a 34–50% reduction under the low concentration of H_2O_2 while they increased by 0%–116% under the high concentration of H_2O_2. The consistent change trend of bacterial percentage and DOC removal indicates that bacteria in cluster 3 might contribute to DOC removal changes between the 0.25 mg/L group and the 0.5 mg/L group (Table 2). In particular, *Zoogloea* spp. (OTU 12210 and 19872) which has a strong ability to degrade different organic materials and has an important function in biological water treatment (Xia et al., 2014) was dominant in the control group, 0.25 H_2O_2 mg/L group and 5 H_2O_2 mg/L group, therefore deserving further consideration as an explanation for DOC removal change.

5. Conclusions

- The increase of DOC degradation with increasing H_2O_2 dosage was caused by a biological process and not by a direct reaction with H_2O_2. The low H_2O_2 concentration (0.25 mg/L) limited DOC biodegradation by 10%, whereas the high H_2O_2 concentration (3 and 5 mg/L) promoted DOC biodegradation by 8% and 28%.
- Low H_2O_2 concentrations (0.25 mg/L) did not influence microbial activity while high H_2O_2 concentrations (1, 3 and 5 mg/L) decreased microbial activity by 23%, 37% and 37%, respectively.
- The bacterial communities in sand were dominated by *proteobacteria*, more specifically, *Betaproteobacteria* (33%–39%). Both 0.25 and 5 mg/L H_2O_2 residuals were proven to influence bacterial community structure. The bacterial community became more diverse after the addition of 0.25 mg/L H_2O_2 but conversely became less diverse when the H_2O_2 dosage increased to 5 mg/L.
- Aerobic bacteria showed different responses to H_2O_2, either sensitive or tolerant. Anaerobic bacteria were found to be sensitive to H_2O_2, and their growth was limited by both 0.25 and 5 mg/L H_2O_2 (17–88% reduction).
- The increased DOC removal at higher H_2O_2 concentrations could potentially be explained by the aerobic bacteria in cluster 3, since microbial activity decreased at low H_2O_2 dosage whereas it increased at high H_2O_2 dosage. The dominant species in this cluster were *Zoogloea* (OUT 12210 and 19872) in the control, 0.25 mg H_2O_2/L and 5 mg H_2O_2/L groups; therefore these bacteria deserve further consideration as an explanation for DOC removal change.
- In conclusion, special attention should be given to the effect of AOP residuals on microbial ecology before introducing AOPs as pre-treatment to biological (sand) processes. In addition, the guideline on the maximum allowable H_2O_2 concentration should be properly evaluated.

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Appendix A. Supplementary data

Supplementary data related to this article can be found at http://dx.doi.org/10.1016/j.chemosphere.2017.07.073.

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