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Removal of natural organic matter using self-assembled monolayer technology

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ABSTRACT

The use of nanotechnologies in water treatment for the removal of natural organic matter (NOM) is a relatively new concept. Using simple self-assembled monolayer (SAM) techniques, a silica substrate was modified as an adsorbent and tested for its potential for reduction of water quality parameters such as UV absorbance, colour and dissolved organic carbon (DOC). Silica particles that were coated with an amino-siloxane SAM (NH_2-SAM) were evaluated in both a high surface area powder form and also a more realistic granular sand form. Initial results using direct stirred contact with powdered NH_2-SAM showed promising results with 60% reduction of UV_{254} after 1 h and up to 70% removal of DOC with higher doses and contact times. NH_2-SAM powder removed NOM in a broader and less selective molecular weight (MW) range than coagulation treatment and this removal was enhanced by pH control at 6, especially for medium MW components. When NH_2-SAM sand was applied, the significantly reduced effective surface area resulted in lower DOC removal but colour removal was still considerable for realistic treatment plant contact times. Attempted regeneration with acidic solutions showed greater effectiveness at lower applied pH, however recovery of adsorption capacity reduced with successive adsorption/regeneration cycles highlighting the need for further refinement of operating conditions for more effective application of this relatively simple water treatment technology.

Keywords: Natural organic matter; Self-assemble monolayer; Water treatment

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1. Introduction

As a result of the demand for improvement of drinking water quality, considerable effort has been made in drinking water treatment research to develop methods to improve natural organic matter (NOM) removal from water. Reduction in the level of NOM before disinfection can minimise the formation of disinfection by-products and reduce the disinfectant residual required to control bacterial regrowth in the distribution system. Conventional water treatment, employing coagulation/flocculation, sedimentation and filtration, has been the most common method for drinking water. However, the use of inorganic coagulants such as aluminium or iron based salts remove only a portion of NOM [1–6]. With increased focus on regulatory requirement for reduction in the level of disinfection by-products in drinking water, there is a need to improve current treatment methods. Research and development into innovative treatment processes is therefore necessary.

Over the past years, nanotechnology has gained a lot of attention and the biomimetic approach for the prepara-

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tion of these materials and devices has attracted considerable interest worldwide, because of its easy handling and considerably lower technical expenditure [7–11]. An example of the biomimetic approach for the synthesis of materials is the use of organic self-assembled monolayers (SAMs) to modify the substrate surface so as to promote the growth of adherent ceramic material [11]. A SAM is a close packed, highly ordered array of long chained hydrocarbon molecules, anchored to the substrate by covalent bonds. The SAM is deposited simply by immersion of the substrate into a dilute organic solution of the hydrocarbon, X–$(CH_2)n$–Y, where Y represents the 'bonding group', such as trichlorosilyl (–$SiCl_3$). X denotes the 'surface group', chosen from among a number of possible species, such as sulfonate (–SO_3H), thioacetate (-$SCOCH_3$), hydroxyl (–OH), amine (–NH_2), nitrile (–CN), methyl (–CH_3), and carboxylate (–COOH) [11], so as to initiate and help sustain the formation of the oxide material when the SAM-coated substrate is transferred to an appropriated ceramic precursor solution (Fig. 1).

Nanoparticles have a high surface-to-volume ratio of atoms and consequently a large fraction of atoms at the surface which provides a high specific surface and unique electronic properties of the nanoparticles. Using this technique, oxide thin films have been synthesized in which the quality, in terms of density, depends on various parameters, such as the concentration of the precursor solution. Generally, the substrate material onto which SAM is deposited is a single crystal of silicon. The trichlorosilyl groups of the surfactant molecules react with the hydroxyl groups on the surface oxide of the silicon and with each other to form a robust, covalently bonded and cross linked siloxane network that anchors the SAM to the substrate. This attachment withstands subsequent exposure to strong acids at temperatures up to at least 80°C, which are the conditions used to deposit the subsequent surface functional groups.

The aim of this study was to evaluate the application of SAM for the removal of NOM. Two base substrates, silicon powder and quartz sand, with different specific surface areas were used to evaluate removal of NOM from water from a South Australian reservoir, Hope Valley. General water quality parameters, colour, UV absorbance at 254 nm (UV_{254}) and dissolved organic carbon (DOC) as well as a more advanced organic characterisation method, high performance size exclusion chromatography (HPSEC), were applied to study the organic matter before and after treatment for the identification of the removable and non-removable (recalcitrant) components of the NOM.

2. Materials and methods

2.1. Preparation and characterisation of SAM coated silica powder and quartz sand

20 g of silica powder (amorphous silicon dioxide, SiO_2, analytical reagent, Malinckrodt Chemical Works, grain size range: 3–90 μm with d_{10} = 7.6 μm, d_{50} = 24.3 μm, d_{90} = 49.1 μm, surface ratio = 0.23 m^2/g) was mixed with 200 mL piranha solution (70% H_2SO_4, 30% H_2O_2 aq. (30 vol. % in H_2O)). The piranha solution oxidizes contaminants and produces free –OH groups on the surface of the powder. After 5 min contact, the suspension was washed with Milli-Q water. Then the powder was filtered with a glass frit filter (pore size 4). Washing was stopped when the pH of the wash water reached the same pH as the applied Milli-Q water. The powder was then dried at 65°C and suspended in 196 mL of anhydrous toluene and 4 mL of surfactant (3-aminopropyltrimethoxysilane) under nitrogen at room temperature. Then the powder was further washed with ethanol (EP grade) for 5 min to remove toluene then followed by Milli-Q water wash to remove the ethanol and dried at 65°C. The SAM coated silica powder is called NH_2-SAM powder hereafter.

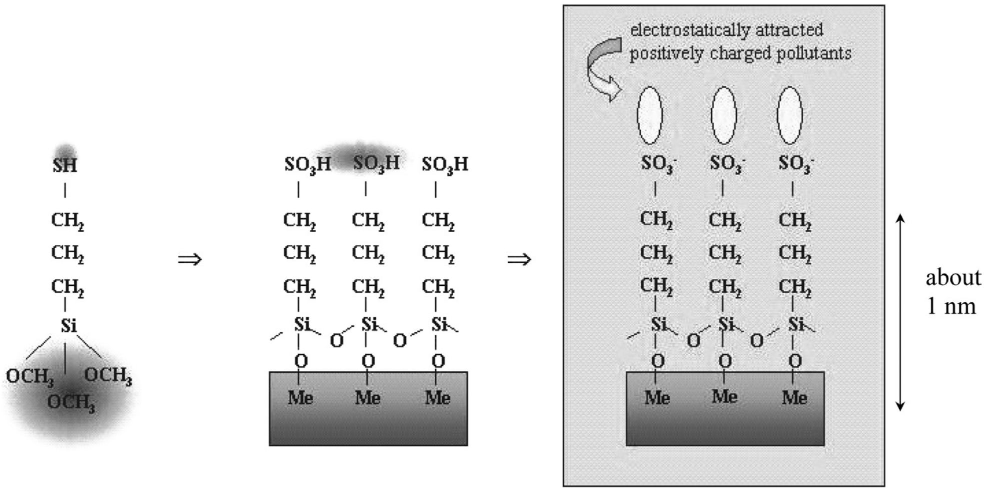

Fig. 1. Formation of a sulfonate SAM on a solid substrate. Me: metal/metal oxide.

The same preparation procedure was used to produce SAM coated quartz sand (crystalline SiO_2, grain size range: 100–450 μm with d_{10} = 121.9 μm, d_{50} = 239.3 μm, d_{90} = 330.5 μm, surface ratio = 0.02 m^2/g). The SAM coated quartz sand is called NH_2-SAM sand hereafter. Physical characterisation of the grain size (both NH_2-SAM powder and NH_2-SAM sand) was determined using a particle sizer (Mastersizer, Malvern, UK).

2.2. Source water for water treatment experiment

The water used for this experiment was collected from the Hope Valley Reservoir (approximately 10 km north-east of Adelaide, South Australia) which supplies water to the Hope Valley Treatment plant. The water is received from the Torrens River system via the Millbrook and Kangaroo Creek Reservoirs which is primarily supplied by the River Murray. The water in this reservoir generally has low colour, medium DOC concentration and high turbidity. The plant employs conventional treatment processes (coagulation/flocculation, sedimentation and filtration processes) and uses alum ($Al_2(SO_4)_3 \cdot 18H_2O$) and a cationic polymer for coagulation.

2.3. Analytical methods

2.3.1. General water quality parameters

DOC concentrations of filtered (0.45 μm) samples were determined using a total organic carbon analyser (Model 820, Sievers Instruments Inc., USA). UV_{254} was measured at 254 nm using a UV/VIS spectrophotometer (Model 918, GBC, Australia) with a 1 cm quartz cell. Colour was determined as described by Bennett and Drikas [13] and measured using a UV/VIS spectrophotometer (Model 918, GBC, Australia) with a 5 cm cell.

2.3.2. Apparent molecular weight determination — high performance size exclusion chromatography

The samples were first filtered through a 0.2 μm membrane filter. HPSEC was analysed using a Waters Alliance 2690 separations module and 996 photodiode array detector (PDA) at 260 nm (Waters Corporation, USA). Phosphate buffer (0.1 M) with 1.0 M NaCl was flowed through a Shodex KW802.5 packed silica column (Showa Denko, Japan) at 1.0 mL/min. Apparent molecular weight was derived by calibration with poly-styrene sulphonate (PSS) molecular weight standards of 35, 18, 8 and 4.6 kDa (Polysciences, USA).

2.4. Treatment experiment

A simple checking procedure (pH check) using Milli-Q water on the NH_2-SAM powder and NH_2-SAM sand (new batch) was conducted to confirm that a good NH_2-SAM surface was formed without leaching of contaminants (no pH change) prior to use for the treatment experiment.

2.4.1. Dose rate/contact time/pH experiment using NH_2-SAM powder

A series of experiments was conducted using a dose rate of 1 g of NH_2-SAM powder with 10 mL of water samples at different contact times (1, 6 and 24 h) and pH conditions (natural — without adjustment and pH 6). The samples were loaded onto a mechanical shaker with speed set on low until the end of the experiment (contact time). Then the experiment was extended to cover various dose rates. Treated water samples were analysed using HPSEC, DOC and UV_{254}. In addition, the experimental procedure was repeated after a regeneration process using either pH 5 or pH 3 solutions. The regeneration process is required to be conducted in an acidic environment, two pHs were selected for comparison.

2.4.2. Using NH_2-SAM sand in column mode with regeneration study

25 g of NH_2-SAM sand was placed into a filtration column (modified from a normal laboratory burette). 300 mL of water sample (adjusted to pH 6) was passed through the column with flow rate adjusted to 0.3 mL/s. Treated water was collected in aliquots of 50 mL. Each aliquot (50 mL × 6) was analysed by HPSEC, DOC, UV_{254} and colour. The procedure was repeated after in-situ regeneration of the NH_2-SAM sand. The NH_2-SAM sand in the column was washed (regenerated) with 50 mL of regenerant (pH 3) followed by 2 × 50 mL Milli-Q water.

3. Results and discussion

3.1. Characterisation of NH_2-SAM powder and sand

One of the objectives of this evaluation is to assess the application of this new concept as a treatment process for NOM removal. In order to provide a comprehensive evaluation, it is important to assess the feasibility of its practicality as a treatment option. Surface area is the most important physical property and generally maximising the surface area, such as using a fine powder, is desirable for more efficient removal. However, the grain size affects manual handling of the adsorbent and is also important in determining the practicality of the process in full-scale drinking water treatment.

Two forms of NH_2-SAM materials were examined, powder and sand. The physical characteristics of both the silica powder and quartz sand were determined using a Mastersizer. The surface area of the silica powder and quartz sand were found to be approximately 0.2 m^2/g and 0.02 m^2/g, respectively. The dose rates were then standardised as surface area per volume of sample (m^2/L) and this standardised unit allows results to be comparable for both forms of NH_2-SAM surface.

The powder has a larger surface area per unit weight and is more suitable for small scale applications such as point-of-use (POU) devices. The NH_2-SAM sand has a small surface area to weight ratio which reduces the removal efficiency but is more practical with respect to manual handling and settled bed permeability and could therefore be applied in larger scale treatment processes such as municipal treatment plants.

3.2. *Removal of natural organic matter*

The DOC concentration of the Hope Valley source water is typically around 6 mg/L. The Hope Valley treatment plant generally removes 30% of this DOC using conventional alum coagulation, sedimentation and filtration treatment train [6]. From previous experience in NOM removal using alum coagulation, improved DOC removal can be obtained by controlling the coagulation pH and generally, pH at 6.2 is an optimum balance between effective DOC removal and minimisation of dissolved aluminium residual in the treated water [6].

3.2.1. *Using NH_2-SAM powder*

The initial optimisation trial was carried out to evaluate the effect of pH on NOM removal using SAM-NH_2 powder. UV_{254} was measured and compared at two different pHs, natural-without adjustment (pH 8) and pH 6 after 1 h of stirred contact between the NH_2-SAM powder (20 m^2/L) and the raw water. The selection of pH 6 was based on the experience of NOM removal by coagulation, however, this also served another purpose that a lot of treatment plants are practicing enhanced coagulation with coagulation pH adjusted at 6. When considering SAM is applied in conjunction with coagulation, to assess the removal effectiveness at pH 6 would be most appropriate. The UV_{254} reading of the raw water was found to be 0.130 cm^{-1}, while water samples after 1 h contact were 0.053 cm^{-1} (60% removal) and 0.070 cm^{-1} (46% removal) at pH 6 and pH 8, respectively. This confirmed removal of NOM is improved at lower pH conditions (pH 6).

A full optimisation trial was conducted to study the effect of contact time and dose rate on DOC removal with pH controlled at 6. From the dose response curves of residual DOC concentrations with different contact time, it was found that the percentage DOC removal increased with higher doses but began to level off towards the plateau at 10 m^2/L of NH_2-SAM powder applied (Fig. 2). The maximum DOC removal was found to be approximately 70% after 10 h of contact. Corresponding DOC analysis of the water samples after 6 h contact time are listed in Table 1.

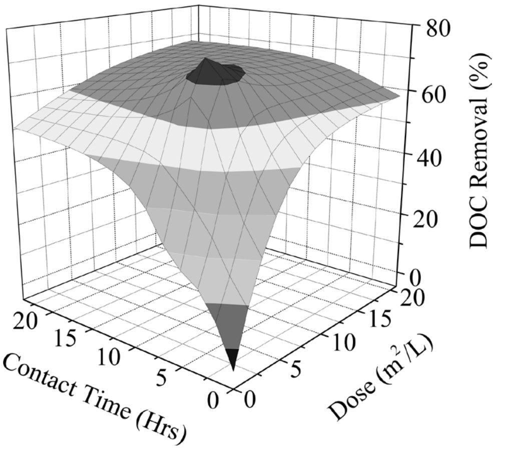

Fig. 2. The surface plot of the relationship of dissolved organic carbon (DOC) removal with dose and contact time using NH_2-SAM powder at pH 6.

Table 1
Dissolved organic carbon (DOC) removal using NH_2-SAM powder at pH 6 with 6 h contact time

	DOC (mg/L)
Hope Valley raw	5.7
After 1 m^2/L SAM treatment	4.2
After 2 m^2/L SAM treatment	2.2
After 4 m^2/L SAM treatment	1.1
After 10 m^2/L SAM treatment	1.0

In a NOM removal study, both the concentration and character are important. HPSEC is a simple characterisation technique for NOM which separates the constituents based on a differential permeation process, according to molecular weight (size). The molecular weight profiles can be used to indicate differences in NOM character. It has proven to be a useful technique for evaluating various water treatment processes by comparing NOM profiles before and after treatment [4,5,14–17]. In Fig. 3a, the Hope Valley raw water has two distinctive profiles, 100–10,000 Da and around 100,000 Da. The latter is an early eluting fractions, i.e. the peaks above 50,000 Da. The composition of NOM responsible for these high MW fractions is thought to comprise colloidal material, however, the reason this peak remains undefined in molecular weight is due to the fact that the molecular weight exclusion limit of the column used in the separation is 50,000 Da. Therefore, this peak can only be used to indicate the presence of colloidal organic materials. This peak is easily removed by a coagulation process [17,18] and this fraction can also be removed by the NH_2-SAM process (Fig. 3). This fraction usually cannot be removed by other adsorption media such as activated carbon [19] or ion exchange resins [20].

The molecular weight region of most interest is between 100 and 10,000 Da. Alum coagulation results in the general removal of the higher molecular weight aromatic organic compounds in this region but there is evidence that compounds below 800 Da are largely recalcitrant for removal by alum coagulation [6,16]. The molecular weight profiles shown in Fig. 3a indicate that the NH_2-SAM process operates with a different removal

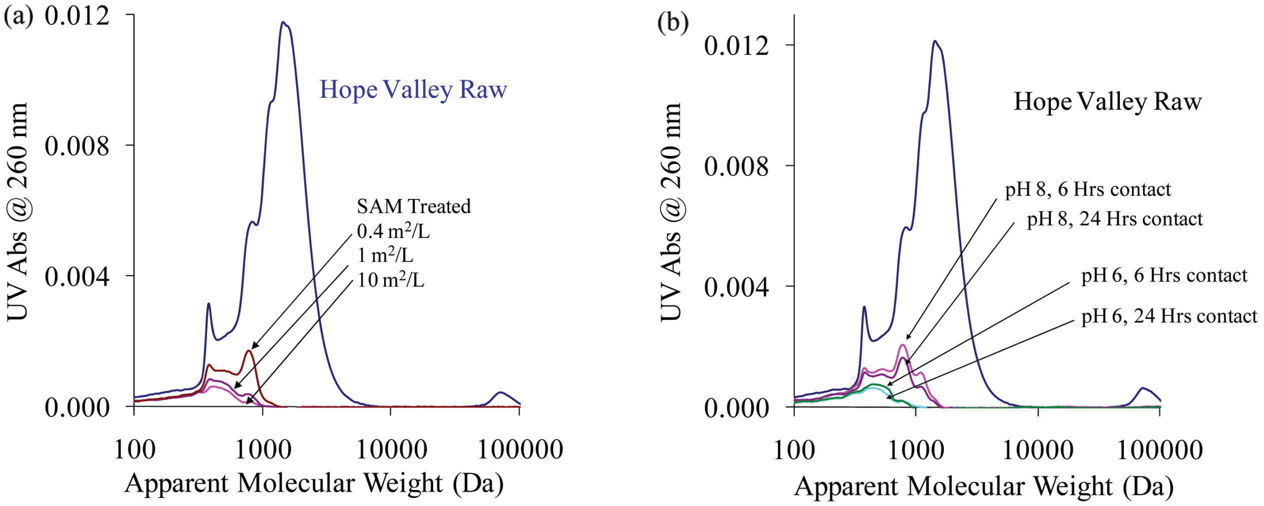

Fig. 3. Molecular weight distribution (HPSEC scans) of raw and treated water samples after NH_2-SAM treatment with different experimental conditions (a) different doses at 6 h contact and (b) at a fixed dose of 20 m^2/L with different pH conditions and contact times.

mechanism to coagulants with a general removal of high MW compounds but, in addition, with higher dose rates, there is also a reduction in the lower MW compounds. Unlike coagulants, the NH_2-SAM appears to adsorb NOM over a broader spectrum of the detectable molecular range. This demonstrates the potential of the NH_2-SAM process as a polishing step after conventional treatment to remove the lower MW fraction which is recalcitrant to alum treatment.

As reported earlier, better NOM removal measured as UV_{254} was achieved at pH 6 compared with pH 8. It was also considered important to determine the character of the organic compounds which were removed under the two pH conditions. The molecular weight distribution profile of two sets of experimental results, pH and contact time, are presented in Fig. 3b. By comparing the chromatograms of the water after NH_2-SAM treatment at pH 6 and pH 8, an additional portion of high molecular weight compounds can be removed at pH 6 as compared with pH 8. The comparison of different contact time shows that extending the contact time to 24 h only marginal improved NOM removal indicating that the adsorbent was already close to saturation after 6 h contact.

3.2.2. *Using NH_2-SAM sand*

A similar optimisation trial for contact times and dose rates was conducted using NH_2-SAM sand (Fig. 4). Due to the smaller surface area to weight ratio compared with the powder, the effective dose rate (m^2/L) of the NH_2-SAM sand is in a lower range compared with the powder. This was reflected by the relatively smaller percentage removal

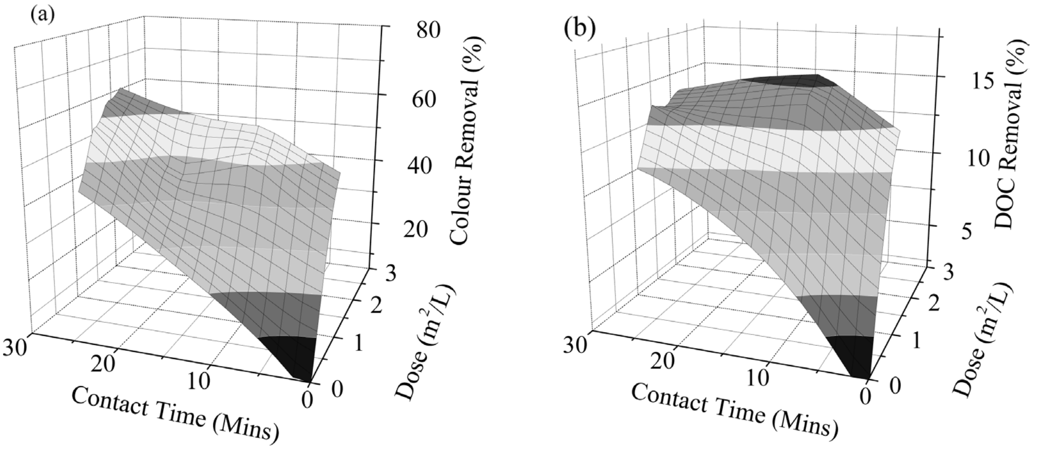

Fig. 4. The surface plot of the relationship of (a) colour (b) dissolved organic carbon removal with dose and contact time using NH_2-SAM sand at pH 6.

of the two parameters, colour and DOC. Ideally, coating NH_2-SAM onto the treatment plant filter sand surface is a very practical way to apply this technology within a drinking water treatment process. However, the removal as compared with powder (larger surface area to weight ratio) is considerably disadvantaged with large doses and long contact times required. The maximum DOC removal in general reduced from 70% to 15% in similar experimental conditions when comparing NH_2-SAM powder and sand, respectively. While the DOC removal is less than might be achieved with virgin granular activated carbon (GAC) filtration, it is comparable to longer term biological activated carbon (BAC) [21], with the advantage of the capacity to easily regenerate the NH_2-SAM sand with pH adjusted backwashing, as described in section 3.3 below.

Generally, colour removal is easily achieved using a coagulation/flocculation process. From the colour removal result (Fig. 4a) using SAM sand, a maximum of 60% removal can be achieved, however this equates to less than 15% removal of DOC (Fig. 4b).

3.3. Regeneration of NH_2-SAM powder

One of the critical considerations of any new potential treatment process is the cost of implementation and operation. While production cost of the NH_2-SAM materials can be greatly reduced by mass production but it is more desirable and practical to regenerate the NH_2-SAM materials. In this trial, a regeneration process was performed using acidic solutions at pH 5 and pH 3. These were used to regenerate NH_2-SAM powder that was used in the previous experiment. To evaluate the effectiveness of the regeneration, the powder was subjected to three sequential adsorption and regeneration cycles and the removal ability of UV_{254}, colour and DOC compared. A summary of the treated water quality and percentage removal of UV_{254}, colour and DOC for Hope Valley raw water is presented in Table 2.

The results shown in Table 2 demonstrated that NH_2-SAM does recover some adsorption capacity using the acidic conditions described above. By comparing all three water quality parameters, the regeneration conditions are clearly more effective at pH 3. From the percentage removal of each parameter, there is evidence of the degradation of the removal capacity after each regeneration indicating that the adsorption is not entirely reversible using the applied regeneration method. The removal of colour was found to be the best out of the three water quality parameters. The regeneration was less effective for recovery of DOC removal capacity, except for the pH 3 regeneration where DOC results contrasted with UV_{254} and colour removal. The reasons for this were unclear and could not be explained within this investigation.

3.4. Treatment experiment using NH_2-SAM sand in column mode

Several application aspects, including physical characteristics, removal performance and regeneration, have been discussed in the previous sections. This section will concentrate on the applicability of using NH_2-SAM coated sand as a water treatment application. The experiment conducted in column mode with in-situ regeneration is based on the conditions obtained and reported previously. 300 mL of Hope Valley raw water was passed through the column at a set flow rate and treated water samples were collected at the outlet of the column in 50mL aliquots to evaluate the removal capacity and saturation point.

The results in Fig. 5 show DOC and colour results of virgin NH_2-SAM sand and in-situ regenerated NH_2-SAM sand. The overall result indicates that the removal performance is reduced in column mode compared with the stirred direct contact mode reported earlier. DOC removal (Fig. 5a) was approximately 10% for the first 50 mL of sample; however no further removal was apparent in subsequent 50 mL samples. This indicated that the kinetics of adsorption were relatively fast, however, the removal capacity is quickly exhausted. A similar result

Table 2
Treated water quality and percentage removal of UV_{254}, colour and DOC for Hope Valley raw water after NH_2-SAM powder treatment (1 h contact time and pH 6) and regeneration in 3 cycles

	UV_{254} (/cm)	% removal	Colour (HU)	% removal	DOC (mg/L)	% removal
Hope Valley raw	0.141	—	17	—	7.1	—
pH5 regeneration						
1st run	0.105	26	10	41	7.2	0
2nd run	0.126	11	13	24	7.0	2
3rd run	0.134	5	15	12	7.1	1
pH3 regeneration						
1st run	0.061	57	2	88	6.5	8
2nd run	0.070	50	3	82	5.7	19
3rd run	0.091	36	7	59	5.8	19

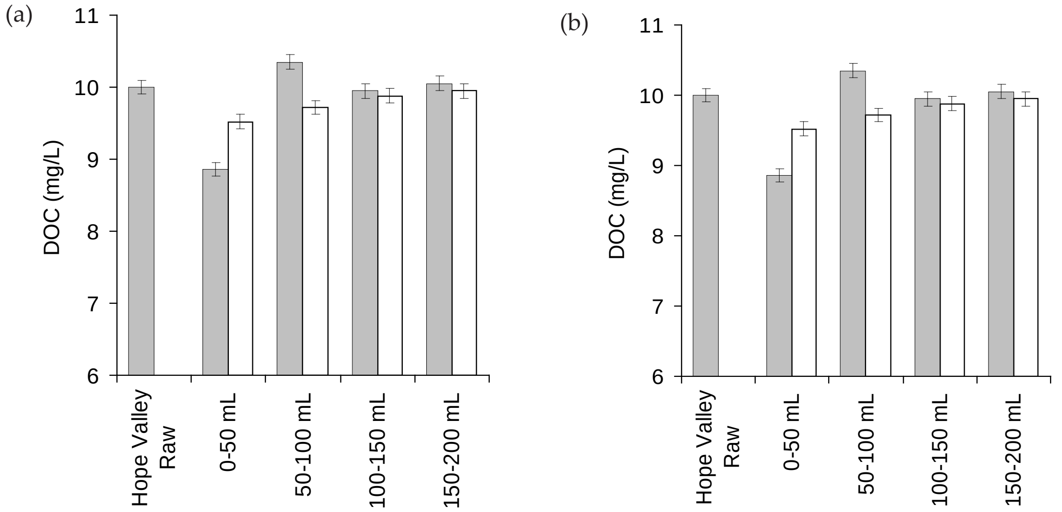

Fig. 5. The treated water quality of Hope Valley raw water after NH_2-SAM (sand) treatment used in column mode with regards to (a) DOC and (b) colour. The first column (shaded) represents virgin NH_2-SAM and the second column (solid) represents the NH_2-SAM after in-situ regeneration.

was observed for colour removal (Fig. 5b). Further development is required to improve the adsorption capacity for this process.

4. Conclusion

Silica particles that were coated with an amino-siloxane SAM were evaluated in both a high surface area powder form (0.23 m^2/g) and also a more practical granular sand form (0.02 m^2/g). Initial results using direct stirred contact with powdered NH_2-SAM showed promising results with 60% reduction of UV_{254} after 1 h and up to 70% removal of DOC with higher doses and contact times. HPSEC demonstrated that NH_2-SAM powder removed NOM in a broader and less selective MW range than coagulation treatment and this removal was enhanced by pH control at 6, especially for medium MW components. When NH_2-SAM sand was applied, the significantly reduced effective surface area resulted in lower DOC removal (approximately 15%) although colour removal was still considerable at 60% for a realistic treatment plant contact time of 30 min. Using an immobilised bed column contactor, the need for greater NH_2-SAM sand to sample ratios were highlighted with 25 g of adsorbent becoming saturated after contact with only 50 mL of a 7 mg/L DOC surface water. Attempted regeneration with acidic solutions showed greater effectiveness at lower applied pH, however recovery of adsorption capacity reduced with successive adsorption/regeneration cycles highlighting the need for further refinement of operating conditions for more effective application of this relatively simple water treatment technology. In terms of application practicality, this material can be applied in two modes, 1) in stirred reactor mode, similar to PAC application at the head of the plant with sufficient contact time and then removed by the coagulation/flocculation process or 2) surface coated onto the filtration media and similar to the normal sand in the filter bed.

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