Aloe sp. leaf gel and water glass for municipal wastewater sludge treatment and odour removal

Short title

Municipal wastewater sludge treatment and odour removal

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Abstract

Aloe gel (Alg), which is a natural extract from the *Aloe sp*. plant, was evaluated in this study for its potential use as a bioflocculant to treat urban wastewater sewage sludge. The gel was used alone and combined with water glass, (WG) under controlled conditions in laboratory experiments. Alg was found effective to settle the flocculated sludge rapidly and remove distinctive unpleasant odours of the sludge as highlighted by GC/MS analysis. Furthermore, Alg was pH tolerant and had no effect in changing the pH of the wastewater. The optimum dose of Alg was 3% at which a sludge volume index (SVI) of 45.4 ml/g was obtained within 30 minutes settling time. To enhance the treatment performances of Alg, WG was also evaluated as an alkali agent to further reduce the chemical oxygen demand (COD) and ammonia (NH4-N) in the wastewater. At equal doses of 3% of WG and Alg each, the combined treatment outcomes showed high turbidity and NH4-N removals of 83 and 89%, respectively but the overall COD removal was at best 25%. The settling rate of treated sludge with combined Alg/WG was very rapid giving an SVI of 25.4 ml/g within only 5 minutes.

Keywords: sewage sludge; *Aloe* sp. gel; water glass; turbidity; COD removal; odour removal.

Introduction

In municipal wastewater treatment, sludge treatment accounts for about a third of the total wastewater treatment plant costs (Nowak 2006), making it one of the important treatment sections of the process. New cost-effective and environmentally friendly sludge treatment technologies are hence needed by wastewater undertakers. In Tunisia, where this study was carried out, the activated sludge process (AS) is the most used biological process for wastewater treatment and the solid-liquid separation of the sludge from the treated water remains a challenge due to overload resulting in poor quality of treated wastewater (Jemli et al. 2015). To enhance the performance of the secondary clarifiers, addition of organic and/or inorganic flocculants is widely practiced. The coagulants/flocculants used in wastewater to aid the separation of sludge can be either inorganic such as aluminium sulphate, or chemically synthetic organic flocculants such as polyacrylamide derivatives (Zahrim et al. 2010). Although these coagulants/flocculants have been successfully used for many decades in the water and wastewater sectors, there have been concerns about their environmental impacts, cost and the sustainability of over-extraction of raw materials, which are not renewable resources. The leaching of monomers from sludges treated with synthetic organic polymeric flocculants has also been highlighted as a significant barrier for the use of wastewater sludge in other areas such as agricultural applications (Abdelaal 2004). Therefore, demand for new coagulants/flocculants to improve the characteristics of the produced sludge to be safely disposed off or utilized in other sectors (e.g. land spraying) is increasing. In recent years, bioflocculants have emerged as potential substitutes to traditional inorganic and synthetic polymeric coagulants/flocculants because they are effective in treating water, generally nontoxic, harmless, less sensitive to pH changes, and are biodegradable (Giri et al. 2015; Liu et al. 2015; Liu et al. 2010). Bioflocculants such as *Moringa* seeds (Menkiti and Onukwuli 2010), *Lablab purpureus* peels (Shilpa et al. 2012), Moroccan cactus extract (Abid et al. 2009), dates seeds (AL-Sameraiy 2012) and *Opuntia dillenii* (Nougbode et al. 2013) have been investigated for turbidity removal providing removal percentages in the range 78 to 94 %. *Moringa oleifera* seed, maize and chitosan were also used in direct filtration of lake water and were successfully evaluated for turbidity and microorganism removal as reported by Mandloi et al. (2004). A widely reported Indian grown natural coagulant (Nirmali seeds), Okra seeds pod tips, sap, plant stalk, and roots have also been studied (Al—Samawi and Shokralla 1996). Al-Samawi and Shokralla (2006) have used okra extract to treat clay suspensions and have concluded that the okra extract was a powerful polyelectrolyte coagulant whether it was used as a primary and/or as a coagulant aid with alum. They also confirmed that the natural okra extract performed much better than alum at high turbidity waters. *Aloe* vera has also been used in water and wastewater treatment to remove suspended solids, turbidity, chemical oxygen demand, heavy metals and textile dyes (Adugna and Gebresilasie 2018; Lee et al. 2015). *Aloe* leaves are well known for their mucilaginous jelly, which is referred to as *Aloe* gel (Femenia et al. 1999; Hamman 2008). *Aloe* gel contains mainly monosaccharides such as glucose and mannose, vitamins, minerals, polysaccharides and phenolic compounds together with some organic acids (Hamman 2008; Mazzulla et al. 2012). In their study on textile wastewater, Adugna and Gebresilasie (Adugna and Gebresilasie 2018) reported that *Aloe steudneri* performed as polyacrylamides for treating the wastewater and suggested that this natural flocculant can substitute the synthetic flocculant. Nougbode et al. (2016) have also confirmed that leaf gels from several *Aloe* species could be used as natural flocculants for water treatment. Adsorption was suggested as the mechanism by which *Aloe* gel provides water treatment due to its high fibre content (Adugna and Gebresilasie 2018). In addition, other constituents of the gel such as glyco-aloe-modinanthrone and tannins are postulated to be responsible for the gel’s coagulation property. Despite being effective to treat water, Aloe gel, similarly to other natural flocculants, could increase the residual organic matter in the treated water. According to literature, the chemical composition of *Aloe* plants largely depends on the species analysed but overall the organic matter can reach up to 81% of the mass of *Aloe* plants (Eugene et al. 2011; Radha and Laxmipriya 2015). In this study, *Aloe* gel was evaluated for the first time as a bioflocculant to enhance the gravity settling of municipal wastewater sludge. The study also highlights the increased performance of using Aloe gel in combination with sodium silicates to obtain faster settling velocities of the treated wastewater sludge. Several jar tests were carried out to select the optimal doses of *Aloe* gel and sodium silicates and the effects on pH changes and the volume of sludge produced.

Materials and Methods

Sludge collection

The sludge samples were collected in two 20 L plastic jerrycans from the wasted line of a secondary settling reactor of an activated sludge process of the municipal wastewater treatment plant (Chotrana II, Tunis, Tunisia). The Chotrana treatment plant serves a population equivalent to 400 000 with a total wastewater flow of 110 847 m^3 per year and serves several industries (textile, slaughterhouse and Food wastewater). The average solids concentration in the sludge is 33 g/L. The sludge samples were immediately stored after collection in a fridge at 5°C. In certain experiments, the pH of the sludge was adjusted using 1M NaOH and 1M HCl solutions.

Preparation of *Aloe* sp. gel

The *Aloe* sp. leaves were harvested in March 2018 from a two-year-old *Aloe vera* plant grown in the garden of the Biotechnology Center of Borj Cedria, Soliman, Tunisia. The gel was prepared as described by Kasmi et al. (2018). Briefly, the whole leaves were washed with a tap water and the spikes which were placed along their margins, were removed before slicing the leaf to separate the skin from the filet. The resulting filets were mixed and homogenized using a hand electric blender (Moulinex, model genuine). The gel was used freshly in each experiment immediately after its preparation.

Aloe sp. gel analysis

Aloe gel composition in terms of fats, sugars, proteins and minerals contents was determined. Fats assessment was performed according to the methodology reported by Hamman (2008). Sugars were assessed using Dubois protocol (Dubois et al. 1956). Proteins content was determined using Bradford methodology (Bradford 1976). As for minerals, phosphorus and magnesium concentrations were determined by means of colorimetric method using vanadomolybdic complex and atomic absorption. Calcium concentration was determined using EDTA method; zinc, copper, and iron assessments were performed using atomic adsorption method (Rodier et al. 2009). Sodium and potassium contents were determined by the flame photometer method.

Preparation of water glass

Water glass (i.e. sodium metasilicate (Na_2SiO_3)) was prepared by reacting Tunisian silica sand (SiO_2 > 99%) with sodium carbonate, Na_2CO_3 (>99%, Honeywell) in a 1:1 M ratio at temperatures between 1200 and 1300°C as indicated by Bouaoun et al. (2017).

Coagulation and flocculation process

The coagulation and flocculation tests were carried out in a jar apparatus equipped with four steel paddles. The doses of the *Aloe* gel varied according to the values 1, 3, 5 and 7 % (v/v) of sludge. The coagulation experiments were performed using a modified protocol as described by Patil and Hugar (2015). The coagulation step was done at high speed of 200 rpm for 2 min followed by the flocculation step at 50 rpm for 30 min. The settling time was set to 60 min after flocculation.

Settling experiments were made using a glass column (46 cm high and 7.8 cm in diameter) in which the height of the liquid/sludge interface was recorded at regular time intervals (Zodi et al. 2009). The effect of pH on sludge settleability was also studied using initial pH values of 7, 11, 12 and 13 adjusted by 1M NaOH solution.

Flocculating rate measurement

To calculate the flocculating rate of *Aloe* gel, Kaolin clay was used to make suspensions at 5 g/L in which, Alg was added and stirred for 2 min. After settling for 1 min, the absorbance at a wavelength of 550 nm of the supernatant sample was measured by a spectrophotometer UV/VIS Lambda 25 (PerkinElmer). The flocculating rate was calculated according to Equation (**1**) (Liu et al. 2010), where A_1 is the absorbance of the supernatant sample at 550 nm and A_0 is the absorbance of the control at 550 nm.

$$\text{Flocculating rate} = (A_0 - A_1)/A_0 \times 100\% \quad \mathbf{(1)}$$

Analytical methods

The solution suspended solids (SS) and volatile suspended solids (VSS) were measured according to the standard methods described by Rodier et al. (2009). The total organic carbon (TOC) was measured using a total carbon analyzer Shimadzu TOC 5050. The total nitrogen and phosphorous contents were determined following the standard methods proposed by Rodier et al. (2009). The pH, conductivity (mS/cm) and total dissolved solids (TDS) (g/L) of each sample were determined using a multi-parameter instrument (C860, Consort bvba, Belgium). Chemical oxygen demand (COD) values were measured by the potassium dichromate colorimetric method using an open reflux system (Rodier et al. 2009). Ammoniacal nitrogen was determined according to the NF T90-15 method (AFNOR). Sludge settling ability is expressed by means of the sludge volume index (SVI) which is often recommended for the characterization of the sludge formation (Kallel et al. 2017; Tchobanoglous et al. 2003; Zodi et al. 2009). The SVI is defined by Equation (2).

$$\mathrm{SVI} = \frac{\mathrm{H_{30}}}{\mathrm{H_0 SS}}\, 1000\ (\mathrm{ml/g}) \quad (\mathbf{2})$$

where H_{30} is the sludge height after 30 min settling (cm), H_0 the initial height of the sludge in the settling column (cm) and SS is the initial sludge concentration after treatment (g/L).

GC–MS analysis VOCs

Volatile organic compounds (VOCs) were analyzed using a headspace (TELEDYNE TEKEMAR HT3TM) coupled with an Agilent GC–MS system (GC with 7890A, mass detector 5975C with Triple-Axis, insert XL MSD). A 30 ml headspace vial was used for incubation of the sample performed during 30 min in headspace oven at 50°C, then transferred in a heated line at 85°C to avoid condensation of VOCs and injected in the GC inlet for 2 min. A HP-5 ms (5 % phenylmethylsiloxane) column (Agilent 19091S-433: 2169.66548) was used (30 m X 250 μm X 0.25 μm) at 1.6 ml/min flow with helium N60 (99.9999 %) as carrier gas and the run was performed over 25 min. The temperature programme of the oven was: 70°C for 2 min, then, 230°C for 20 min and 270°C for 25 min. The Inlet had the following characteristics: temperature of 250°C, helium gas carrier, split flow of 20 ml/min, splitless split/splitless inlet: splitless injection mode, 60.688 kPa as inlet pressure, 50 ml/ min and 2 min for purge flow and time, respectively; gas saver on and 20 ml/min for gas saver flow and 2 min for gas saver time. In this analysis, the used MS had the following specifications: ms quadrupole at 150°C, from 50 to 550 m/z full scan, 70 eV ion ionized energy, source and transfer line temperature at 250°C. The volatile compounds were identified by reference to mass spectra and the retention time of Wiley09 NIST2011 library.

Results and Discussion

Sludge characterization

The main characteristics of the collected sludge are summarized in **Table 1**. The results reveal that the mean value of 7.15 for pH, 11200 mg O_2/L for COD and 16 g/L for SS. VSS content was 2.6 g/L, the TOC was about 473 mg/kg and the ammonical nitrogen and phosphorus contents were 400 mg/L and 6.53 mg/kg respectively. Compared to a similar sludge reported in Ramirez et al. 2018, we observed a low carbon-nitrogen content of Tunisian sludge and quite high level of COD which might be attributed to the industrial effluent co-treated at the Chotrana wastewater treatment plant.

Aloe sp. gel characterization

In order to provide meaningful discussion on the effect of the bio-flocculants on the sludge treatment, the *Aloe* sp. leaf gel characteristics were determined (**Table 2**). The analysis revealed an important amount of soluble sugars of about 22.8 g/100g of lyophilized gel and high content of proteins (7.8 g/100g). Fats minerals and metals were also detected.

Treatment of sludge with *Aloe* gel and water glass

Effect of Alg and water glass addition on sludge's pH

Fig.1 illustrates the effect of the *Aloe* sp. gel (Alg) addition on the sludge's pH. The figure also shows the effect of adding water glass (WG) alone or combined with Alg (Alg-WG) on pH values. Following the addition of 3 mL/100mL Alg, the pH showed only a modest increase from 6.95 to 7.9 to remain constant at 7.9 regardless of the increased amount of Alg added. However, the addition of 3 mL/100mL water glass exhibited a remarkable increase in pH from 6.95 to 12.5. A further increase in WG has increased pH modestly to reach a value of pH 13 after adding 7 mL/L of water glass. In fact, the alkaline character of sodium silicate is the main cause of pH increase as shown by the reaction equation:

$$Na_2O.\ SiO_2 \xrightarrow{H2O} 2Na^+ + SiO_3^{2-}$$

Effect on turbidity removal

Fig.2 shows the turbidity changes versus the added volumes of Alg and WG separately and in combination. The results show that as the added volume increases, the turbidity decreases. According to Fig 3, Alg was less effective than water glass to remove turbidity (Alg: 45%; WG: 89%). This could be attributed to the formation of calcium silicate as main precipitate following the reaction of water glass (sodium silicate) and calcium content in the sludge **(Table 1)**. The obtained precipitate had a potential to adsorb fine particles making the supernatant free of particles.

Effect of Alg and WG addition on SS and VSS

Fig.3 illustrates the effect of the different doses of *Aloe* gel and water glass added separately or in combination on both SS and VSS removal. The obtained results show that the SS content increased instead of being reduced. However, no significant effect was noticed for VSS removal with both treating agents used separately or combined. The progressive addition of WG in the sludge at contents higher than 1% induced a reestablishment of the SS content to its original value (~20.3 g/L) after a drop to 14.5 g/L at 1% WG (**Fig. 3b**). In order to investigate the combined effect of *Aloe* gel and WG addition on the sludge treatability, further experiments were carried out at fixed Alg content of 3% while WG dosages were varied from 1 to 7%. The more WG was added, the higher the SS content was recorded (ranging from 20.4 g/L at 1% WG dosage to reach 27.5 g/L at 7% WG). The VSS values recorded a moderate increase from 9 g/L at 1% to 10 g/L at 7% **(Fig. 3c**).

Effect of Alg and WG addition on COD and NH_4-N

Fig. 4 illustrates the changes of COD and ammonia nitrogen concentrations with the added volume of *Aloe* gel and water glass separately and combined. At low doses, the recorded data reveal a slight increase of COD value when aloe gel was added to the sludge (**Fig. 4a**). Similar observations were also reported by Ramavandi and Farjadfard (2014). This increase in COD following the addition of Alg was expected due to the organic matter content of *Aloe* gel which is rich in organic substances (e.g. carbohydrates and proteins). Yet, above 1mL/100mL of added Alg, the COD values of the solution showed a decreasing trend but the overall COD of the solution remains higher than the initial COD of the sludge until the applied dose of Alg exceeded 5 mL/100 mL Alg. A further increase in Alg doses above 5 mL/100 mL, results in COD values slightly lower than the original COD value of the sludge. In contrast, the addition of WG alone or combined with Alg resulted in a much more effective COD removal than Alg reaching a removal percentage of about 25% at a dose of 7mL/100 mL water glass.

Fig. 4b shows the removal of NH_4^+-N at different doses of Alg and WG. According to Fig. 5b, the addition of Alg and WG alone or combined at low doses resulted in a significant reduction in ammonia. At a dose of 1mL/100 mL, the removal of NH_4^+-N was 66% for all agents to reach 89% at 7mL/100 mL WG. A further increase in Alg dose above 1mL/100 mL did not increase NH_4^+-N removal but it made it worst (only 40% removal at 7mL/L), possibly due to a competition with COD removal as illustrated in **Fig. 4a** where COD removal with aloe gel becomes relevant only at high doses. In addition, Alg active constituents (e.g. proteins) may aggregate and fold at high doses (Gupta et al. 1998), which reduces the number of active functional groups available for NH_4^+ accumulation and adhesion. The action of water glass was much more pronounced since its alkali character changes ammonium to ammonia gas(pKa=9.25 at 25°C) that can be easily removed from solution via the increase of pH to 12.9 (**Fig. 1**); which justifies the high ammonia removal of 89% obtained following the addition of WG.

Performances of *Aloe* gel and water glass on sludge settling

Fig. 5 depicts the sludge settling performance for Alg and for combined Alg-WG treatments as well as the raw sludge without any coagulant being added. The SVI values are also indicated in **Fig.5**. The combined Alg-WG treated sample exhibited a net drop from 100 mL to reach 15 ml within only 5min. The settled volume stabilized at 10 ml after 10 min until the end of the experiment (90 min). The Alg treated sample settled volume was limited to 28 mL after 40 min or higher settling times. However, it is noteworthy that both Alg and combined treatments recorded better results compared to the settling of raw samples without addition of any of the coagulants; where, at best, a final settling sludge volume of 34 mL was recorded after 80 minutes settling. The effect of the optimal dosages of the combined treatment of sludge after settling using 3% of Alg and 3% of WG dosages showed a net difference of settling rates. **Fig .6** gives the particle size distribution of the untreated sludge, sludge with *Aloe* gel and sludge with both *Aloe* gel and water glass. Particle size shifts are clearly observed for the Alg-WG treated sludge. The sludge particles shifted from fine sizes (~100µm) to larger particles and the agglomerates were found to have a bimodal distribution with a mean size value of 300µm. This might explain the accelerated settling velocity observed in the presence of WG**.** The increased particle size as a result of WG addition could be explained by the formation of large aggregates resulting from the interaction between silicate polymers (i.e. WG) and sludge with possible ion binding with inorganics in the wastewater (e.g. Ca^{2+}) (Okano et al. 2015; Zuo et al. 2015) . The SEM images (**Fig. 7**) show that the structures of the raw sludge and the treated sludge with Alg-WG have an abundance of micro pores. The SEM images suggest that significant changes in the floc's morphology occurred following the addition of Alg-WG as evidenced by the formation of large aggregates showing pillars of 10 to 20 µm of length between aggregates in the presence of Alg-WG. The observed building structure might be attributed to calcium silicate due to the reaction of silicates in the water glass with calcium in the sludge (Okano et al. 2015). The outcome of this novel combined treatment of wastewater sludge by addition of *Aloe* gel and water glass highlighted the clear macroscopic aspect of coarse agglomerates of sludge with raw sludge having suspended fine particles.

Performances of *Aloe* gel on sludge odour removal

The mass spectrum of VOCs emitted by the untreated sludge is illustrated in **Fig. 8a.** The analysis allowed the detection of four compounds identified as Toluene (peak 1), Benzenamine, N-ethyl- (peak 2), 4-fluoro-1,2-xylene (peak 3) and phenol, 2,6-bis(1,1-dimethylethyl) (peak 4) with retention times (min) of 5.087, 12.394, 13.415 and 17.981 and relative areas (%) of 41.03, 38.34, 12.76 and 7.86, respectively. **Fig. 8b** and **c** illustrate the mass spectra of VOCs emitted by the treated sludge using Alg and WG, respectively. The spectra show that the addition of either Alg or WG results in the disappearance of peaks 2, 3 and 4 initially detected among the untreated sludge VOCs. Nevertheless, a new compound identified as D-limonene with retention time (min) of 9.867 and relative area of 48.64% was detected after Alg addition. Limonene is one of the most widespread terpenes in the flavour and fragrance industry (Zodi et al. 2009). A human olfactory sensing experiment was carried out and showed that the twenty people did not detect any unpleasant smell from the treated sludge, which provides a further evidence of the removal of odour following the application of Alg.

Conclusion

This study presented a new insight of the potential use of *Aloe* gel for the treatment of municipal wastewater sludge as a novel bioflocculant. *Aloe* sp. leaf gel, at different doses (1, 3, 5 and 7%), was tested as bioflocculant via sequential treatments that used coagulation, flocculation and sedimentation processes under defined operating conditions. The *Aloe* gel showed good performance for sludge solid- liquid separation. To enhance the *Aloe* gel action, a further step was necessary by the addition of water glass. The combined treatment showed that the use of aloe gel and water glass at 3% yielded a removal of turbidity up to 83%, and 89% of ammonia nitrogen. Moreover, the resulting flocs from the treatment using aloe gel and water glass generate much coarser particles and readily settling sludge, typically with only 5 min of settling time. SEM investigation enabled the observation of multi pillars of 10 to 20 µm length located within the sludge intra particles. Furthermore, the *Aloe* leaf gel odour removal efficiency was revealed through VOCs analysis of the treated sludge which indicated the disappearance of odour-causing substances following application of Aloe gel or water glass. The proposed process would be an attractive approach not only to reduce the treatment cost but also to minimize environmental concerns and generate eco-friendly sludge.

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Figure Captions

Fig. 1 pH values variation following the *Aloe* sp. leaf gel (Alg), water glass and aloe + water glass addition to sludge at different volume.

Fig. 2 Turbidity variation with different volume of *Aloe* sp. leaf gel (Alg), water glass and *Aloe* gel + water glass

Fig. 3 Variation of Suspend Solid (SS) and Volatile Suspended solid (VSS) of sludge following the application of (a) *Aloe* sp. leaf gel (Alg), (b) water glass and (c) aloe + water glass

Fig. 4 Variation of COD (**a**) and ammoniacal nitrogen (**b**) with volume of *Aloe* sp. leaf gel (Alg), water glass and *Aloe* + water glass

Fig. 5 Effect of *Aloe* gel (□) and combined Aloe gel-Water glass (□) addition on the treated sludge compared to the raw sample (□) during 90 min

Fig. 6 Raw and treated sludge particle Master size distribution

Fig. 7 SEM image of raw sludge (**a**), and (b) combined *Aloe* gel and water glass treated sludge.

Fig. 8 Mass spectra of VOCs from the untreated sludge (**a**) and the treated sludge using *Aloe* sp leaf gel (**b**) and water glass (**c**)

Table 1 Characteristics of sludge generated by Chotrana municipal wastewater treatment plant.

Measured parameters	**Unit**	**Recorded values**
pH	-	7.15 (at 25°C)
Moisture content	%	84 ± 1
Dry matter content (SS)	%	16 ± 0.5
VSS	g.Kg^{-1}	2.6 ± 0.5
TOC	mg.Kg^{-1}	473 ± 5
Total nitrogen	mg.Kg^{-1}	33.2 ± 0.5
Total phosphorous	mg.Kg^{-1}	6.53 ± 0.05
COD	mg O_2 L^{-1}	11200 ± 10

Table 2 Aloe leaf gel biochemical characterization

Lyophilized Aloe sp. leaf gel constituent	**Content (g/100g)**
Fats	4.8
Soluble sugars	22.78
Proteins	7.82
Ca	3.02
Mg	0.98
Na	3.03
K	3.89
P	0.01
Fe	0.87
Cu	0.04
Zn	0.01

Figure 1

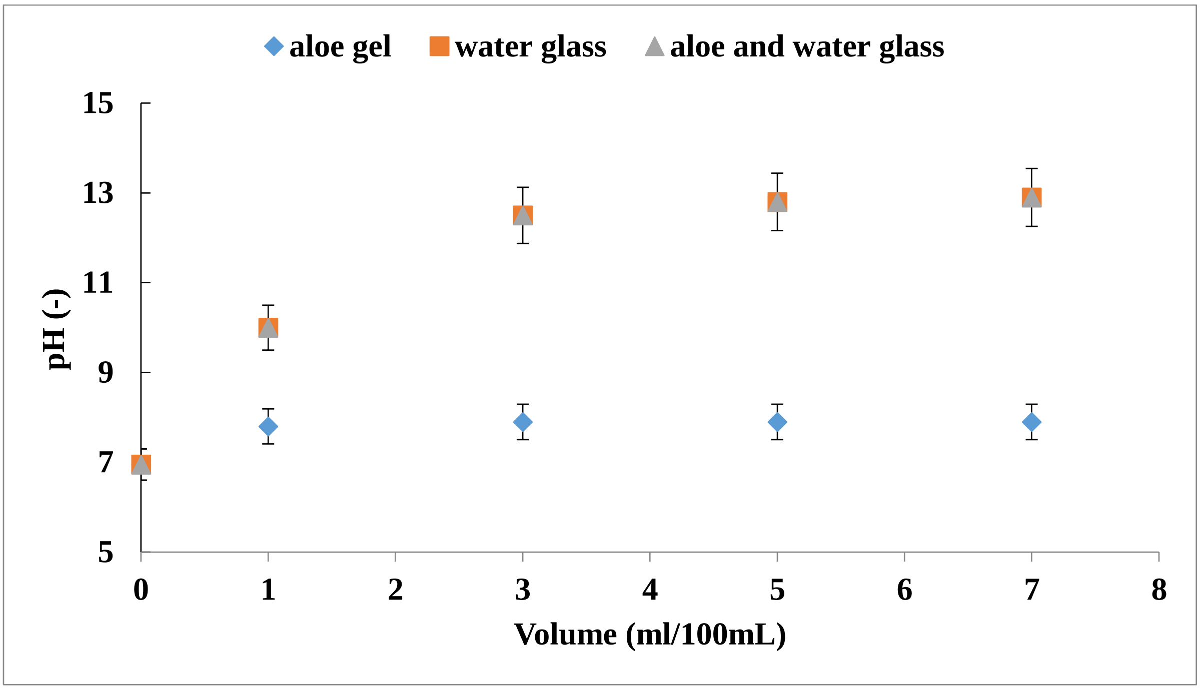


Figure 2

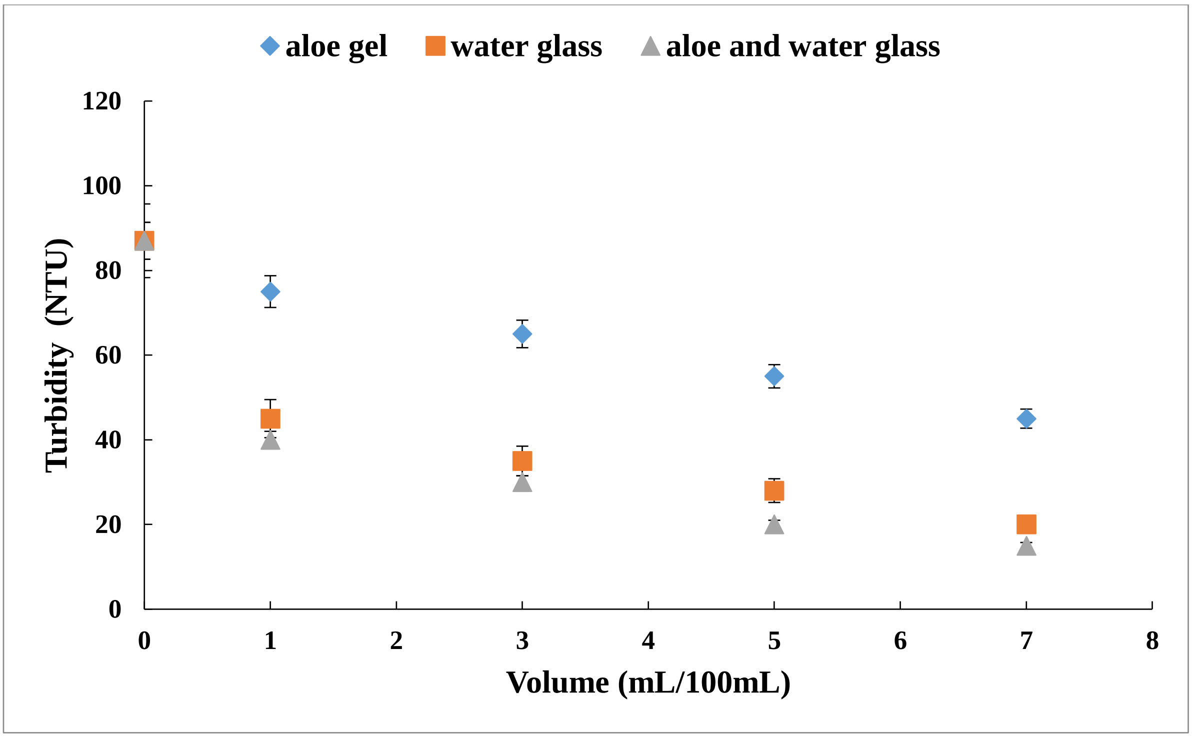


Figure 3

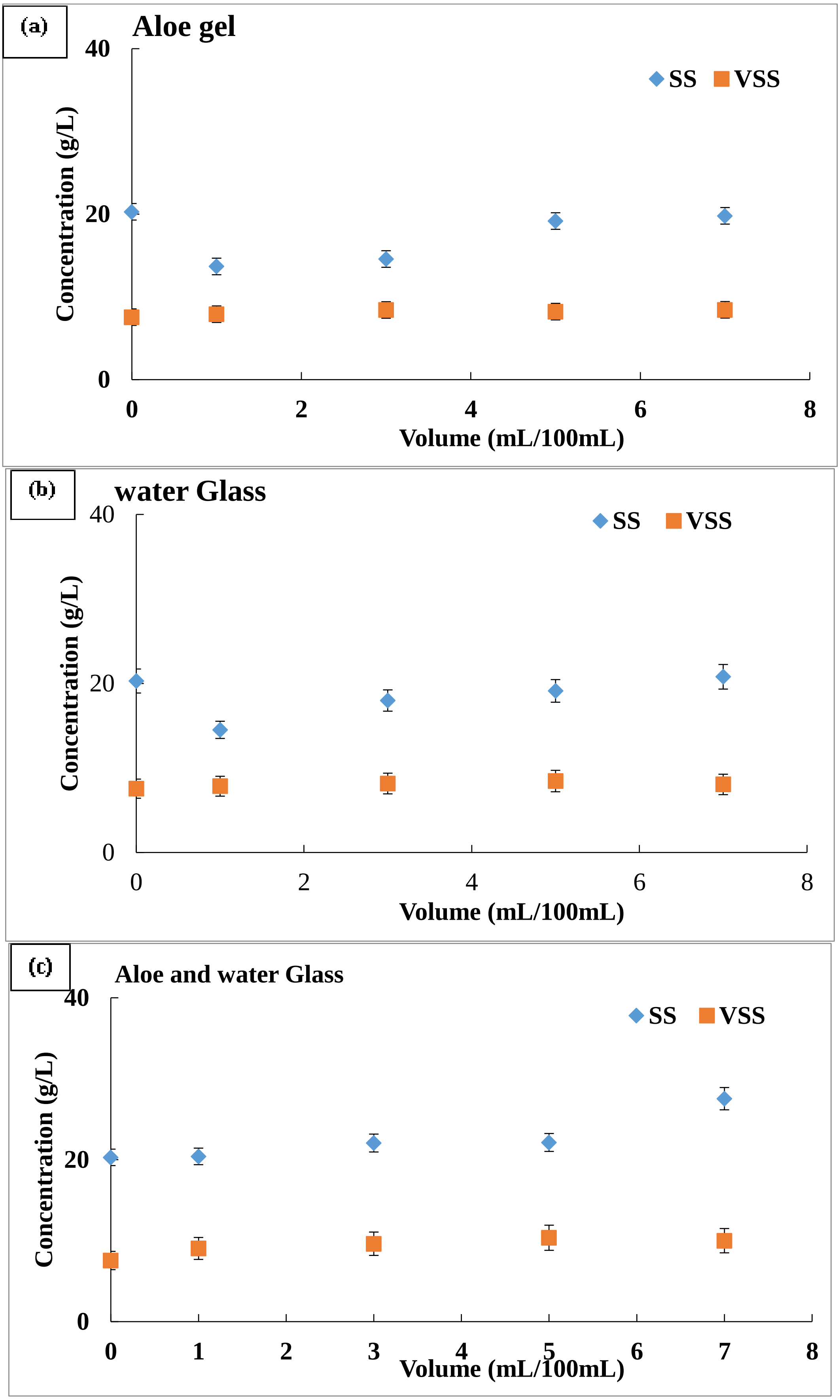

(a)
Aloe gel
SS
VSS
Concentration (g/L)
40
20
0
0
2
4
6
8
Volume (mL/100mL)
(b)
water Glass
SS
VSS
Concentration (g/L)
40
20
0
0
2
4
6
8
Volume (mL/100mL)
(c)
Aloe and water Glass
SS
VSS
Concentration (g/L)
40
20
0
0
1
2
3
4
5
6
7
8
Volume (mL/100mL)

Figure 4

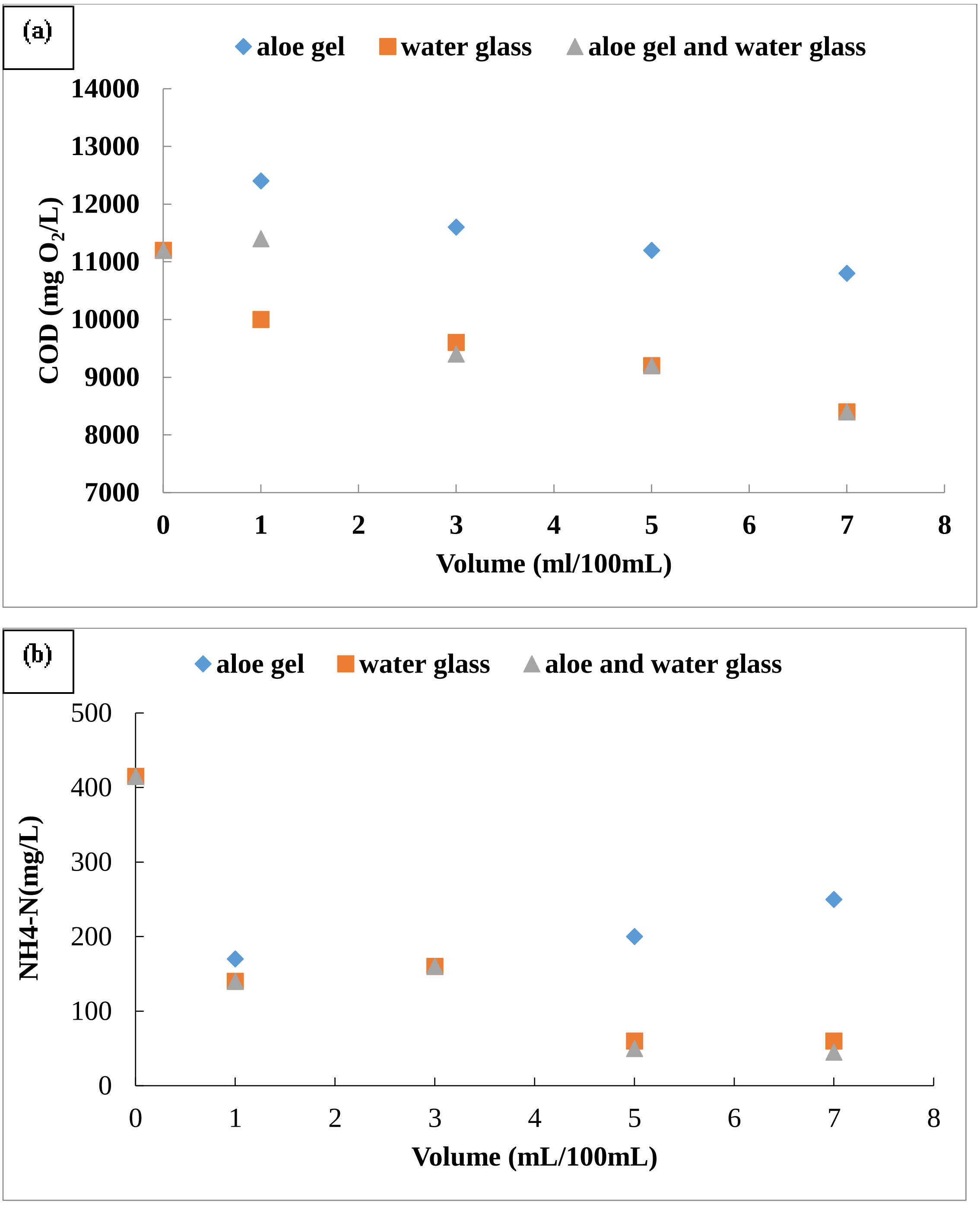


Figure 5

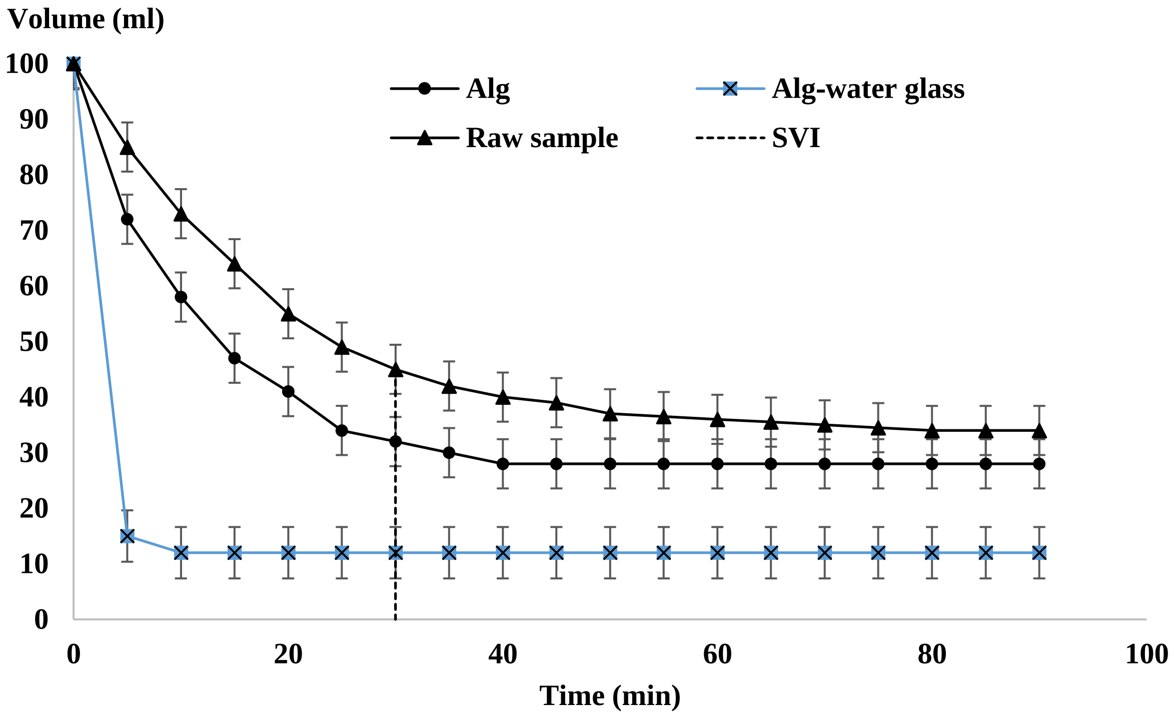

Volume (ml)
100
90
80
70
60
50
40
30
20
10
0
Alg
Alg-water glass
Raw sample
SVI
0
20
40
60
80
100
Time (min)

Figure 6

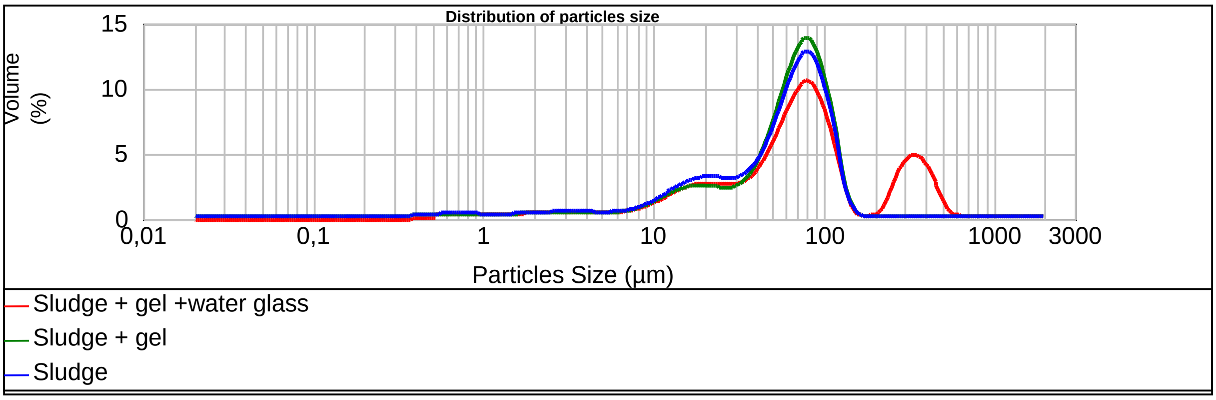

Distribution of particles size
Volume (%)
15
10
5
0
0,01
0,1
1
10
100
1000
3000
Particles Size (µm)
Sludge + gel +water glass
Sludge + gel
Sludge

Figure 7

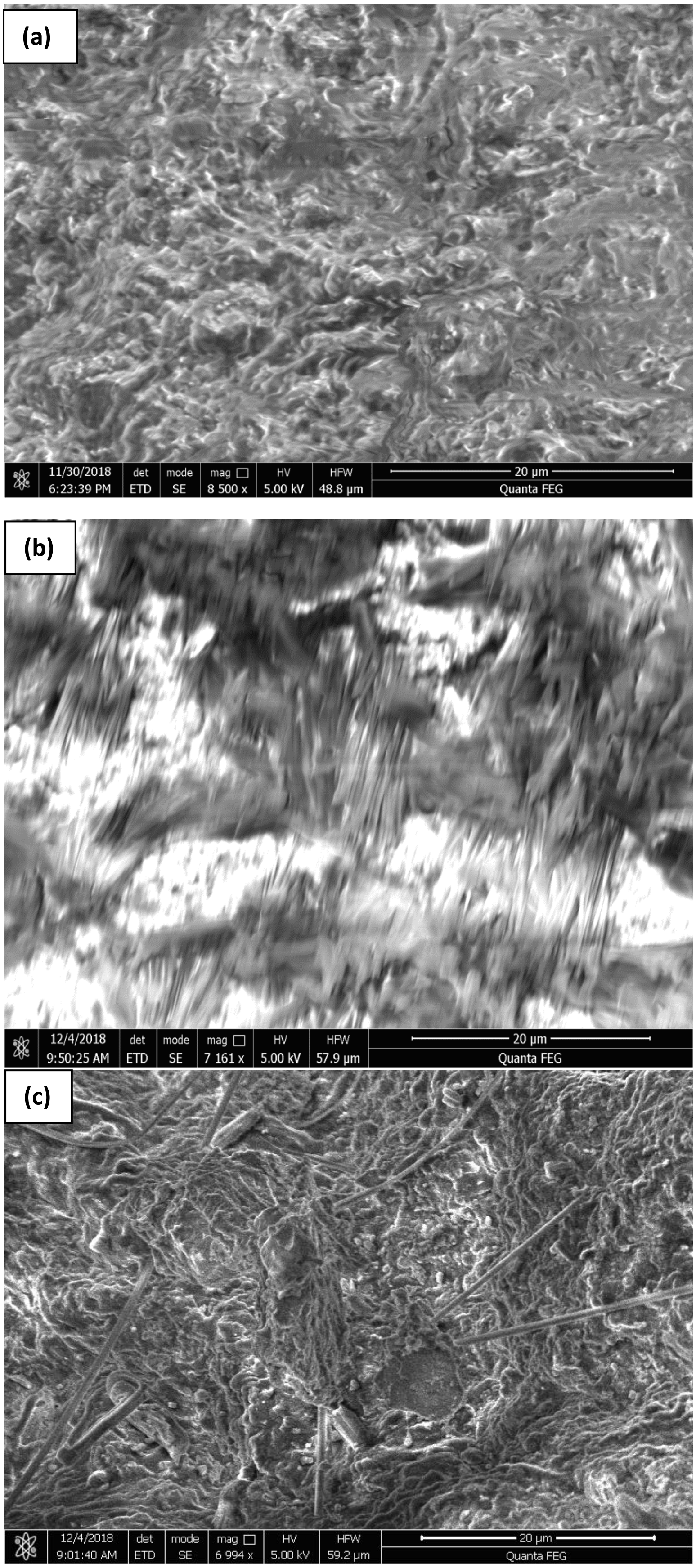
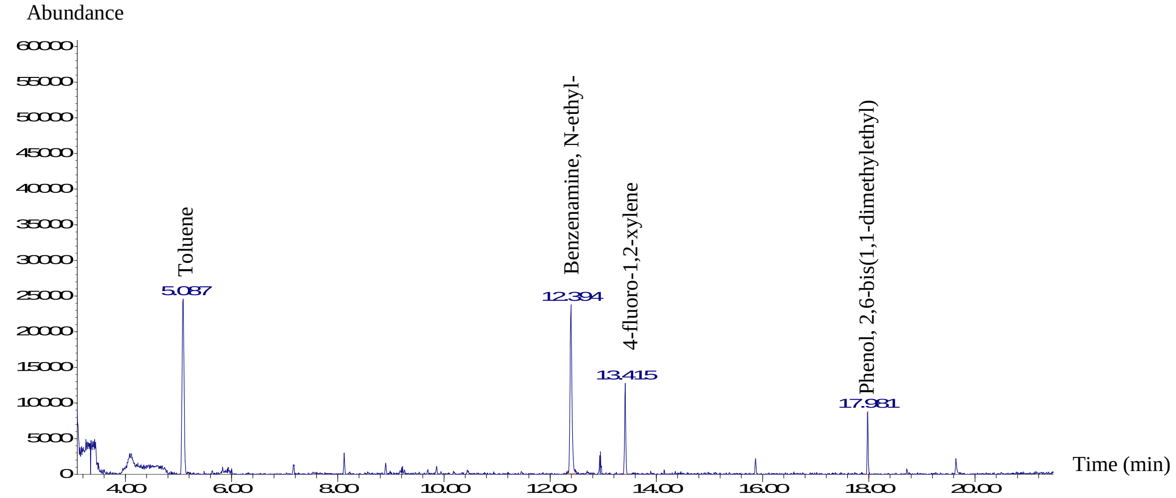
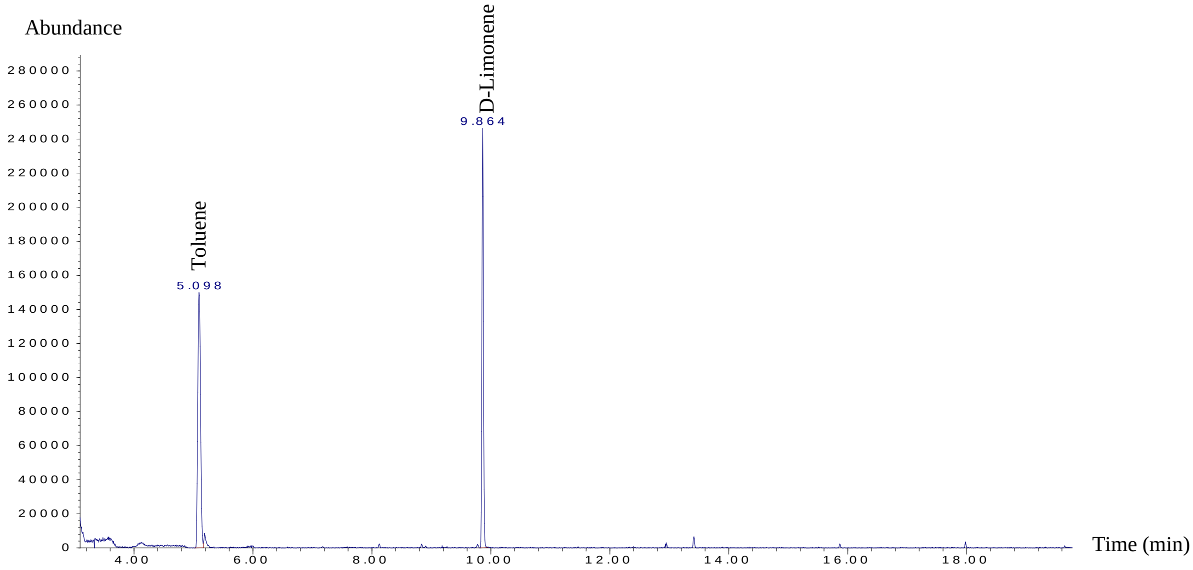


Figure 8

(a)


Abundance
60000
55000
50000
45000
40000
35000
30000
25000
20000
15000
10000
5000
0
Toluene
5.087
Benzenamine, N-ethyl-
12.394
4-fluoro-1,2-xylene
13.415
Phenol, 2,6-bis(1,1-dimethylethyl)
17.981
Time (min)
4.00
6.00
8.00
10.00
12.00
14.00
16.00
18.00
20.00

(b)


Abundance
280000
260000
240000
220000
200000
180000
160000
140000
120000
100000
80000
60000
40000
20000
0
D-Limonene
9.864
Toluene
5.098
Time (min)
4.00
6.00
8.00
10.00
12.00
14.00
16.00
18.00

(c)

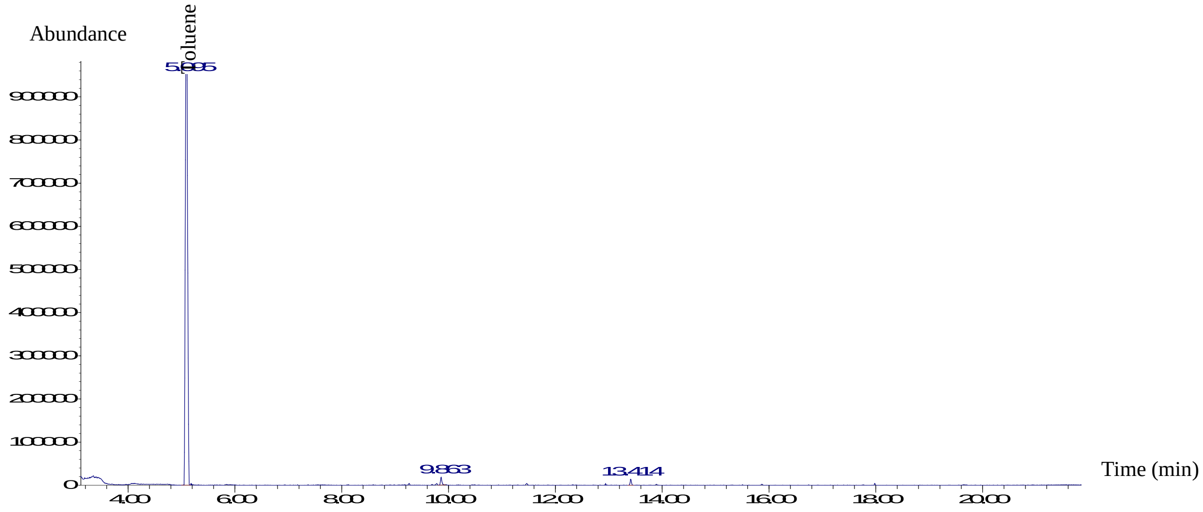

Abundance
Toluene
5.095
900000
800000
700000
600000
500000
400000
300000
200000
100000
0
9.863
13.414
Time (min)
4.00
6.00
8.00
10.00
12.00
14.00
16.00
18.00
20.00

Figure S1: The untreated sludge (**a**) and the treated sludge using the combined *Aloe* gel and sodium silicate at a dosage of 3% (**b**)