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Research article

Technical and economic investigation of chemical scrubber and biofiltration in removal of H₂S and NH₃ from wastewater treatment plant



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ABSTRACT

A detailed techno-economic comparison of a chemical scrubber (CS) and a bio-filter (BF) was conducted over a 45-day time period at a municipal wastewater treatment plant (WWTP), Yazd city. The assessment of emissions quantity indicated that odor emissions from the Yazd WWPT mainly consist of hydrogen sulfide (H2S) and ammonia (NH3). It was also found that odor gaseous loading changes corresponding to water consumption pattern in society ($R^2 = 0.922$) for H_2S and ($R^2 = 0.978$) for NH_3 . The highest level of 25 and 3 ppm for H_2S and NH₃, respectively were detected at specific times during the day. The BF system was continuously supplied with Yazd WWPT's off-gas treatment while the CS was only examined at the times during the day when the gas emissions are at the highest level. The removal efficiency of NH3 and H2S were found to be affected by their respective loading rate. Additionally, among the various oxidants examined in the CS, the NaOCl solution showed the best results in terms of removal efficiency and compatibility. The experiment revealed almost complete removal of NH_3 while the H_2S removal efficiency remained above 95% for both systems regardless of the operating conditions. This study clearly demonstrates the effectiveness of both systems in treating actual waste gases containing H₂S and NH₃. By comparing the gas loading rate of both systems and considering limitations of the BF system, the CS seems to be more efficient applicable odor control technology from a technical viewpoint. From the economic viewpoint, comparisons revealed that chemical usage and operating expenses were costly parts of the CS and the BF, respectively. The economic indexes of $1.58 \, \text{c.m}^{-3}$. h^{-1} and $2.57 \, \text{c.m}^{-3}$. h⁻¹ were obtained for the BF and CS, respectively, reflecting cost-effectiveness of the BF system.

1. Introduction

The development and infringement of urban residential areas on potential malodor sources specifically wastewater treatment plants (WWTP) has led to an increasing number of public grievances against these malodorous compounds (Lebrero et al., 2011). Frequent exposures to these malodors have a direct threat to people's health and welfare (Chen et al., 2001; Cheng and Hsieh, 2010). This factor, along with environmental sticking legislations about air pollution control in connection with industries and odor resources, has led to the need for odor management (Sucker et al., 2009). Odorous compounds are primarily produced during biological metabolisms in wastewater collection and treatment systems (Jaber et al., 2014). Generally, major sources of smell encountered in WWTP are ammonia (NH₃) and

hydrogen sulfide (H₂S) (Alfonsín et al., 2015; Nisola et al., 2009). H₂S and NH₃ are highly toxic, colorless, corrosive and irritating malodorous nuisances with very low odor thresholds. These compounds not only are a direct threat to human health but also represent a remarkable contribution to photochemical smog formation and particulate secondary emission (Gao et al., 2001). Hence, it is essential to control the emission of these gases not only for public health and safety but also for environmental protection using a cost-effective and eco-friendly odor abatement system (Van der Heyden et al., 2015). Up to now, various technologies have been proposed and applied for the treatment of malodorous components from contaminated air which can be classified into physical/chemical (chemical scrubbers, incinerators, adsorption systems, and so forth), biological systems (biofilter, bio-trickling filter (Arellano-Garcia et al., 2012; Gabriel et al., 2013; Qiu and Deshusses,

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2017), bio-scrubber, and activated sludge diffusion reactors) or a combination of them. Chemical scrubbing (CS) and bio-filter (BF) are among the most commonly employed treatment techniques in WWTPs (Kumar et al., 2013; Lebrero et al., 2011).

The CS is a well-known, widely established, and reliable technology. This mainly arises from short gas retention time (as low as 1–2.5), extensive experience in design and operation, high robustness when properly operated, and rapid start-up of the wet scrubber (Moussavi et al., 2008; Vega et al., 2014). Removal efficiencies of up to 98% for $\rm H_2S$ and 99.8% for $\rm NH_3$ are commonly achieved under good operation practices (Lebrero et al., 2011). However, this technique is relatively costly and produces secondary pollutants.

In contrast to physical/chemical treatments, biological process is often considered a cost-effective and eco-friendly route for the purification of gas streams (Van Groenestijn and Kraakman, 2005). The main environmental impact of biological techniques was caused by high water consumption to maintain biological activity. Among microbiological processes, the BF has proved to be one of the most promising methods in WWTPs' odor treatment (Lewkowska et al., 2016). The removal efficiency of an odor gas by a BF is generally satisfactory under appropriate sets of conditions. However, it reduced significantly when applied to undiluted and hardly biodegradable waste gases at large quantities of the contaminated gas stream (Lebrero et al., 2011). The BF is also criticized for high land and material requirements; and their long period of acclimation required for microbial population (weeks or even months) (Lebrero et al., 2011). In addition, treatment of low solubility gases such as NO in conventional biofilters has typically required long contact times (Kumar et al., 2010). Recently, the membrane bioreactor (MBR) technology for air pollution control has been proposed (Kumar et al., 2008, 2009). In spite of operational advantages compared with the biofiltration, membrane bioreactors for air pollution treatment have yet to be tested at full scale and their long-term operational stability still has to be demonstrated. Furthermore, an MBR has a high construction cost disadvantage (Luvsanjamba et al., 2008).

Both of these technologies (BF and CS) have been widely reviewed in the literature, and their optimal range of application and performance for odorous compounds treatment has been clearly established on the basis of laboratory and field experiences. Despite numerous studies conducted on the BF and wet scrubbers, only a few ones have been focused on the comparative evaluation of these odor-treatment technologies (Estrada et al., 2010; Gao et al., 2001). In this regard, Gao et al. compared a BF and a sodium hypochlorite/sodium hydroxide wet scrubber in order to control H2S in the Mill Greek Wastewater Treatment Plant in Cincinnati, Ohio economically and technically (Gao et al., 2001). In another study, various odor control technologies including BF, activated sludge diffusion, bio-trickling filtration, CS, activated carbon adsorption, regenerative incineration, and a hybrid technology have been comparatively evaluated in terms of environmental performance, process economics, and social impact by using the IChemE Sustainability Metrics in the context of odor treatment from wastewater treatment plant (WWTP). The odor pollution from a WWTP located at Stuttgart University was selected as a model of malodorous emission for the environmental evaluation. Methyl mercaptan, hydrogen sulfide and mainly, VOC components were included in the above-mentioned model emission (Estrada et al., 2010). According to this simple literature survey and to the best of our knowledge, no research has dealt with the detailed comparison of the wet scrubber and BF in order to simultaneously remove NH3 and H2S in an air stream. On this basis, a detailed techno-economic comparison of these two odor-abatement technologies was conducted over a 45-day time period at a municipal WWTP (located at Yazd, Iran) as model odor source to provide a basic guideline for technology selection in odor management in Yazd WWTP. In this regard, "removal efficiency" and "total annualized cost" are taken as technical and economic indexes, respectively. In line with technical evaluation, the effects of the gas loading rate and EBRT for both systems and also, oxidant nature and scrubbing flow rate for the wet scrubber,

on simultaneous H₂S and NH₃ removal were also investigated.

2. Materials and methods

2.1. Site description of biofilter and chemical scrubber

Two odor control systems were designed and installed at the Yazd wastewater treatment plant in Yazd, Iran. The treatment plant is an SBR activated sludge treatment facility, which treats municipal wastewater of Yazd city. This study was conducted over the months of June and July 2017 in which odor emissions were at a high level during the year. Both control systems were designed to control off-gases and odors from the pumping station because this section had a great portion of waste gas emission from the plant.

2.2. Chemicals

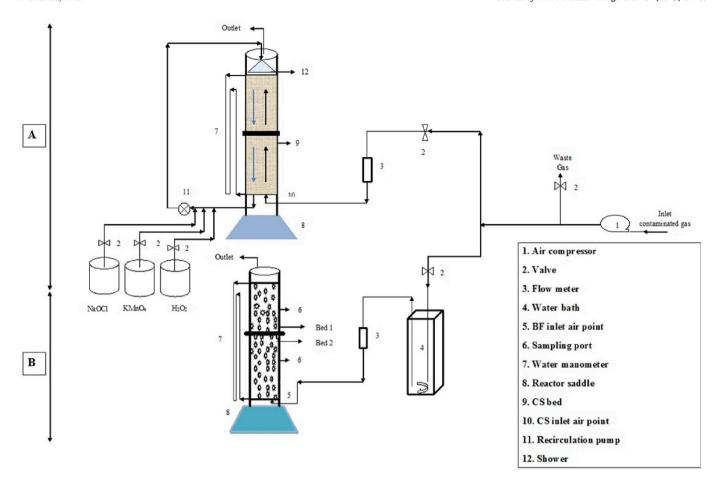
The material used were: N, N-dimethyl-p-Phenylenediamine, H_2SO_4 50%, FeCl₃, and Na₂S.9H₂O that was applied for the determination of H₂S ranges in samples, and Na₂ [Fe (CN) 5NO], NaOH 6.75 g mol⁻¹, NaOCl 0.1 N, C6H5OH 45%, Na₃PO₄.12H₂O, CHCl₃, and (NH₄)₂SO₄, were used for preparation of solutions which were utilized for detection of NH₃. Besides, CdSO₄.H₂O used as a precursor to making alkaline suspension of cadmium hydroxide as an absorption solution for capturing H₂S from polluted air. In addition, H₂SO₄ (0.1 N) was used as NH₃ absorption solution. The oxidant compounds (NaOCl and H₂O₂) purchased in the commercial grade with purities of 12% and 35%, respectively, but KMnO₄ was 99% purity in analytic grade. All the chemical compounds in this study except NaOCl and H₂O₂ were analytic grade (Merck-Germany) and used without further purification.

2.3. Biofilter odor control system

Fig. 1 indicates the experimental setup of BF at Yazd wastewater treatment plant. It was made up of Plexiglas with a height of 95 cm and an internal diameter of 14 cm. BF consists of two sections and a 2 L bottle at the bottom for the collection of the solution. Each section is filled with equal amount of bed material to a height of 40 cm giving a total working volume of 12.03 L. The bed materials (Fig. 2) used for the treatment of real gaseous mixture were activated sludge with concentration of 4-6% containing ammonium-oxidizing bacteria (AOB) to improve nitrification, which mixed with Poly Vinyl Chloride (PVCs), which were first chopped to pieces with 2 cm dimension and afterward, these pieces were cut into three equal parts with using longitudinal incision (approximately 6.6 mm) and a compost (density of $500\,\mbox{kg}\,\mbox{m}^{-3}\mbox{)}.$ The bed materials in each section were supported by plates made of Plexiglas®. The experimental air for the operation is supplied using an air compressor, which was prepared by sucking in the ambient air. The Flow of air into the BF was adjusted using a flow control valve attached to a flow meter (Brooks Instrument Company).

2.3.1. Moisture supply methods in BF system

In fact, we applied two methods for creating moisture for microorganism activity in BF: (1) a water bath, and (2) water trickling from the top of the BF. In the first method, input air was passed through the water bath (diameter of 15 cm and height of 80 cm) before entering the bed. It caused input air loads water vapor droplets, and then these vapor droplets could be dispersed uniformly through the whole of the bed. In the second method, we added 200 mL of the deionized water (pH \approx 7) into the bed about every three days. In low airflow rates, the water bath used to provide moisture, and in high input airflow rates, both methods (1 and 2) were applied to provide moisture in the bed. As regards, in the high input airflow rates, there is a high microorganism activity in the BF bed , the temperature of the system increases further, and conversely, the moisture content decreases. Thus, the supply of moisture in high airflow rates is more difficult.



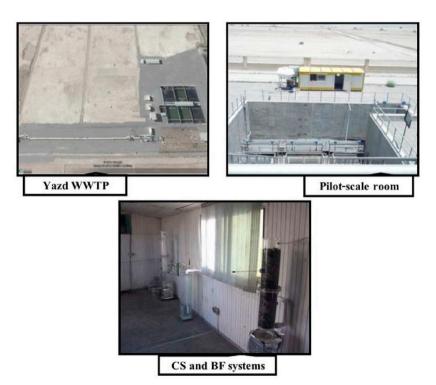


Fig. 1. Schematic flow chart and real pilot of systems, (A) Chemical scrubber, (B) Biofilter.



Fig. 2. Biofilter bed components and preparation.

2.4. Chemical scrubber

The schematic of the CS system is depicted in Fig. 1, the pilot-scale packing bed scrubber was constructed using Plexiglas with an internal diameter of 14 cm and a total height of 120 cm. The bed height and volume were 70 cm and 10.77 L, respectively. The bed material made of Polyethylene (PE) with the porosity of 80% and specific surface area 369.6 m² m⁻³. In this system, the polluted air is supplied by a suction pump, the inlet airflow rate to the CS was adjusted and monitored by a flow meter and provided valves. Three plates were placed across the reactor for holding the packing material and helped to diminish the channeling of the fluid through the bed. The area below the bed served as a liquid pool to save the scrubbing solution for recirculation. A diffusing pump circulated this liquid from the pool and sprayed on the top of the bed using a shower. Three chemical oxidants consisting of NaOCl, H₂O₂, and KMnO₄ were utilized for oxidizing odor mixture compounds. The odor treatment formula in this system was the reactions that happen between circulating liquid and malodorous air in a reverse current in the CS bed. The oxidants, which were made in a basin, had been injected (4–10 L min⁻¹) into the CS bed by a centrifuge pump and a shower.

2.5. H₂S and NH₃ analysis

In order to assess the efficiency of each system, we need to measure the concentrations of malodorous chemicals in the inlet and outlet of the gas.

 H_2S : It was used per the Jacob and et al. the method for measuring H_2S gas. The H_2S sampling set up consisted of two impingers and an air vacuum pump and tubes. A vacuum pump aspirated the air with $Q=1\,L.\text{min}^{-1}$ and $T=5\text{--}30\,\text{min}$ into the impinger that contains $45\,\text{mL}$ adsorption solution of cadmium hydroxide. To protect samples from sunlight, impingers were covered entirely by aluminum foil. Impinger solutions were analyzed using DR 5000 at wavelength $\lambda=670\,\text{nm}$. The H_2S concentration (ppm) in the air was calculated by the following equation (Lodge Jr, 1988).

$$H_2S(ppm) = ((A \times 0.719))/Vs$$
 (1)

Where A is H_2S (µg) in solution, Vs (L) is the volume of air that passes through the adsorbent solution in standard circumstance, and 0.719 is the volume of 1 mg H_2S gas in stoichiometric conditions.

 NH_3 : The NH_3 sampling method resembles H_2S but the Q of the vacuum pump is $1-3\,L\,\text{min}^{-1}$ and $T=30\,\text{min}$ into the impinger that

contains 20 mL adsorbent solution of sulfuric acid. Impinger solutions were analyzed by DR 5000 at wavelength $\lambda = 630$ nm. The NH₃ concentration (ppm) in the air calculated by the following equation (Lodge, 1988).

$$NH_3 (mg.m^{-3}) = ((17.03 \times NH_3 ppm))/24.45$$
 (2)

In here 17.03 is NH_3 molar mass and 24.45 is the molar volume of an ideal gas in 25 $^{\circ}C$ and 101.3 kPa.

2.6. Measurement of moisture, pH, and temperature in biofilter

In order to maintain a suitable level of moisture, every three days roughly 200 mL of deionized water was trickled from top of the BF. Therefore, everyday sampling from two sections of the bed was done and after analysis in the laboratory, the moisture in each bed of the BF calculated by Mn=Mw/Mo

Where Mn is the moisture content, Mw is the mass of medium with water, Mo is the mass of medium without the water. For measurement pH, it took 5 g of each bed substrate, from sampling ports (6) in Fig. 1 and mixed it with 50 mL water, and afterward, this was set up for the time of 30 min on the mixer with a speed of 350 rpm. Finally, the pH of this composite was determined by using an Ecoscan pH meter (Eutech instruments). The temperature of the bed was also measured in three periods of the day (morning, afternoon and evening) by placing a thermometer in each bed of BF.

2.7. The EBRT, airflow rate (Q), pressure (ΔP), and gas velocity measurement in systems

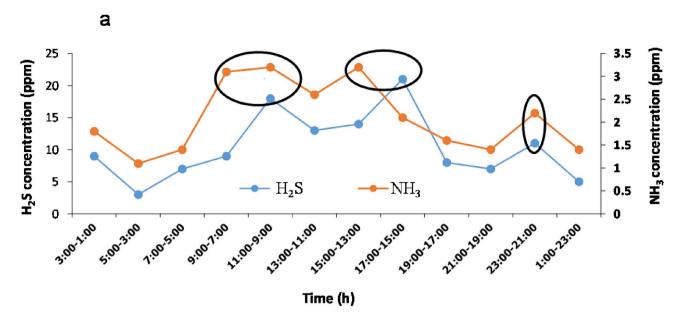
The EBRT and airflow rate (Q) in systems are calculated by the following equations (Eq. (4));

EBRT =
$$Q/V$$
 and $Q = V/t$ (3)

Where Q is the airflow rate (L.min $^{-1}$), V is reactors bed volume (m 3), and t is gas contact time (s). The pressure drop in either the CS or the BF bed was determined by a water manometer, which was installed in two head of the beds and the water level fluctuates in this device before and after sampling for regarding EBRT, calculated ΔP in systems. The gas velocity in systems was estimated by this equation:

$$U_{SG} = Q_G/A \tag{4}$$

Where U_{SG} is gas velocity m.s⁻¹ and Q_G is inlet gas flow rate m³. s⁻¹ and A is the surface area of the bed m².



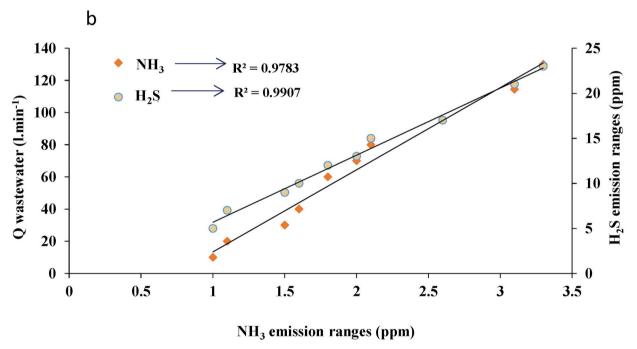


Fig. 3. (a) H₂S and NH₃ daily emissions and those peaks (for 23rd day); (b): Relevance between inlet wastewater to Yazd WWTP with H₂S and NH₃ emission ranges during a day.

Table 1 H₂S efficiency & removal rate for BF.

Time (day)	EBRT (s)	Q air (L/min)	C _{inlet} (ppm)	C _{out} (ppm)	Efficiency (%)
1-9	60	9.2	17.75	2.8	84
12-16	60	9.2	24.02	0.29	99
16-26	30	18.4	20.13	0.54	97
26-38	20	27.6	23.36	0.56	98
38-45	15	36	22.23	1.7	92

2.8. Economical evaluations

The economic analysis must take into account equipment, construction costs, and operating costs (raw materials, utilities, and operator wages). Then, costs including capital and expense of the various options should be compared using economic parameters as net presentworth or annualized cost. In this study, the annualized cost method has been chosen and applied. In the Supplementary Data, details of the cost estimation procedure are depicted (Gao et al., 2001).

Table 2 NH₃ efficiency & removal rate for BF.

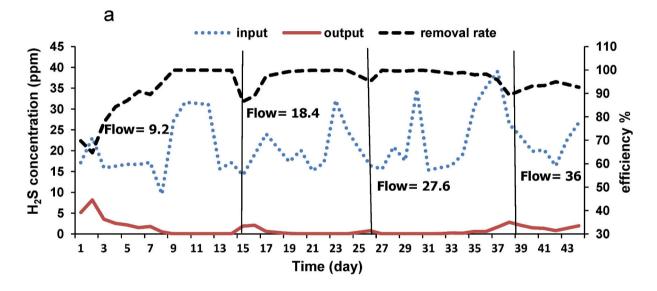
Time (day)	EBRT (s)	Q _{air} (L/ min)	C _{inlet} (ppm)	C _{out} (ppm)	Efficiency (%)
1-6	60	9.2	2.1	0.26	88
12-16	60	9.2	2.71	0.018	97
16-26	30	18.4	2.14	0.037	98
26-38	20	27.6	1.88	0.036	98
38-45	15	36	1.97	0.007	99.6

3. Results and discussions

3.1. H₂S and NH₃ emission ranges

Assessment of the odor compounds level and quantity of emissions is essential for the better performance of odor control systems. This data will also help operators to shut the odor control system down when marginal levels of malodorous compounds are in the environment or

start-up it in times when there are high concentrations of them. Fig. 3a shows daily H₂S and NH₃ emissions at Yazd WWTP in Iran. As can be seen, odor emission ranges are low, except for peak periods. The highest level of the odorant gases observed in time ranges of 9:00-11:00 a.m., 3:00-5:00 pm, and 11:00-12:00 p.m. during the day with the main peak concentration of about 25 and 3 ppm for H₂S and NH₃, respectively. According to the results, it can be seen that there is a direct relationship between water consumption pattern in the community and odor emissions. It is clear that the highest level of odor emissions occur during the peak period of water consumption. With increasing water consumption the $Q_{\text{Inlet wastewater}}$ increases, and this phenomenon leads to an increase in the odor emissions at WWTP. Meanwhile, with this scrutiny, it can be seen that there is a relationship between water consumption and production of NH3 and H2S gases. In general, several factors have a role in the occurrence of sulfur cycle in wastewater networks such as microorganisms in wastewater, wastewater volume, the amount of sediments in the sewer, and wastewater contact area with air (Lens and Kuenen, 2001). The sulfur cycle is completed in four main processes including production of sulfide, transition sulfide from



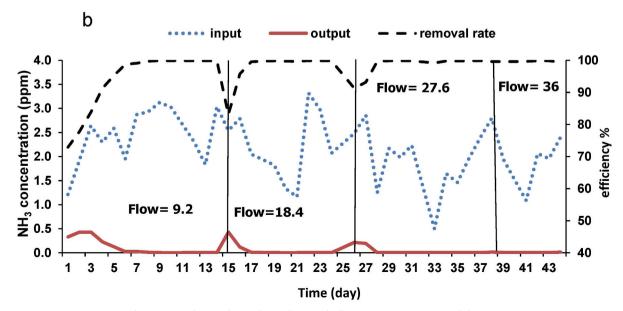
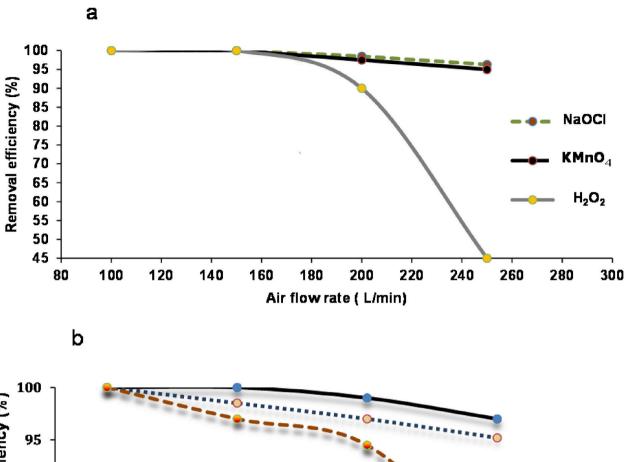


Fig. 4. $\rm H_2S$ and $\rm NH_3$ inlet, outlet, and removal efficiencies in BF. (a) $\rm H_2S$ and (b): $\rm NH_3$.



NH₃ removal efficiency (%) 90 NaOCI KMnO₄ 85 H₂O₂ 80 160 180 200 220 80 100 120 140 240 260 280 300 Air flow rate (L/min)

Fig. 5. H_2S and NH_3 removal efficiencies by CS in present of three oxidants. (a) H_2S and (b): NH_3 .

liquid to gas phase, chemical and biological oxidation of sulfur, and the reaction of sulfide ion-containing compounds with iron and chemical sequestration, sulfide created by bacteria, which converted sulfate into the sulfide (Parande et al., 2006). This bacteria activity occurred in the anaerobic condition in wastewater treatment collection networks and is seen in areas where the wastewater flow moves slowly and is poorly aerated (Hvitved-Jacobsen et al., 2000). Ammonia is another substance that produces odor in the sewer by bacterial decomposition of urea in wastewater networks. Nitrogen organic compounds and urea are converted into ammonia through the ammonification aerobic process in wastewater collection networks. On the other hand, NH3 has a low evaporation temperature, and it can quickly be evaporated and released into the environment (Talaiekhozani et al., 2016). Therefore, H2S and NH₃ are produced when the water consumption rate is low and wastewater flow speed is slow in the sewer, but when this flow goes up in wastewater pipes (high water consumption) these odorants are released into the air. This marked relevance between odor emission rate and

 $Q_{Inlet\ wastewater}$ has been shown in Fig. 3b with the correlation coefficient $R^2=0.990$ and $R^2=0.978$ for H_2S and NH_3 , respectively. This relation is important in the CS start-up times more than other technologies.

3.2. Biofilter efficiency and removal rate study

3.2.1. Effect of gas flow rate and EBRTs in simultaneous removal of $\rm H_2S$ and $\rm NH_3$

The efficient and simultaneous removal of $\rm H_2S$ and $\rm NH_3$ by the biofiltration process as evaluated and the results are given in Tables 1 and 2. The $\rm H_2S$ and $\rm NH_3$ removal process can be classified into four stages in the BF system in terms of inlet air (9.2, 18.4, 27.6 and 36 L. min⁻¹) and EBRT (15, 20, 30 and 60 s) Fig. 5. The incubation time is one of the factors determined when BF has reached a stable state and is robust in the reduction of malodorous compounds. In fact, after this time biological activity starts in the BF bed. In the start-up, these incubation

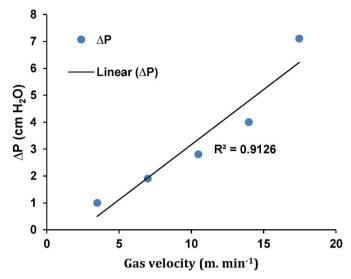


Fig. 6. ΔP fluctuates corresponding to gas velocity (airflow rate) changes in CS.

Table 3
Oxidants fractions in CS.

Oxidant	Consumed oxidant $_{\rm mg}/{\rm Oxidized}$ malodorous $_{\rm mg}$	Scrubbing liquid pH
Cl ₂	6.5–7.1	8–9
H ₂ O ₂	1.5–2.2	7.8–8.1
KMnO ₄	9.5–10.4	7.5–8.1

periods were 9 and 6 days for H2S and NH3, respectively. These incubation figures were partly higher in comparison with other studies mentioned in the literature (Chen et al., 2018; Chung et al., 2000; Galera et al., 2008; Hou et al., 2016; Jones et al., 2004; Kim et al., 2002; Rabbani et al., 2016). Forasmuch as prepared biomass (containing adapted bacteria), was not used in the BF system, so it needed more time to start up biological activity (adaptation) and reach stability in the system. On the other hand, in aerobic conditions, H₂S is converted to sulfate (SO₄)²⁻ by sulfur-oxidizing bacteria (SOB), and the pH ranges for optimal growth of SOB are variable (1.8-7.4), which depends on the type of bacteria. The pH value is 7.4 and below 1 for Thiobacillus denitrificans and Acidithiobacillus thiooxidans, respectively. In the same environment, AOB (ammonia oxidizing bacteria) and NOB (nitrite oxidizing bacteria) convert NH₃ to NO³⁻ and the optimum range of pH for these kinds of bacteria are 6.0-9.0 for Nitrosomonas and 7.3-7.5 for Nitrobacter (Lors et al., 2009; Solcia et al., 2014). According to pH graphs in two beds (Fig. S1b), pH in initial days (around 7.9-7.8) was favorable to NH3 converting bacteria rather than H2S ones, so NH3 degradation by BF was begun sooner than H₂S in the contaminated air. After a stable condition was reached in the BF system, in EBRT = 60 s the removal rate of H₂S and NH₃ were 99% and 97% respectively. Further diminished EBRTs (15s) caused deterioration in the removal ability of BF. Decreasing EBRT increases the ΔP value (from 4 to 6 mm H₂O) and a gradual drying of the biomass materials occurs in the BF bed, following that H⁺ ions increase in the system which created the acidification event in packing materials, as a result, this phenomenon is a hamper to biological activity. Hence, with the reduction of bacteria activities in the BF, the removal efficiency dropped as well (Lebrero et al., 2013; Ma et al., 2006). Therefore, with increasing the airflow rate from $27.6-36 \, L \, min^{-1}$ and EBRT from 20 to 15 s the BF removal efficiency for H₂S decreases from 98 to 92% Fig. 4a. Additionally, increasing airflow rate in the system enhances mass loading, which is mass of contaminant entering the BF per unit volume of filter materials per unit time, and consequently disturbing adsorption processes between bed bacteria and pollutant (Fulazzaky et al., 2014; Lebrero et al., 2013). Thus, with an airflow rate of 36 L min⁻¹ the amount of H₂S that

is loaded in the system increased (maximum 35 ppm) and caused a reduced removal efficiency from 98 to 92%. These factors (ΔP and mass loading) did not have any noticeable effect on the BF performance for the removal of NH₃. This is attributed to this fact that the mass loading rate (maximum 3 g m⁻³. h⁻¹) entering to the system is not so high and the BF is able to degrade approximately all of NH₃ gas (99.6% EBRT of 15s) Fig. 4b. The obtained results are consistent with the results of Rehman et al. (2009) when after a power failure, the maximum removal efficiency of H₂S diminished from 100 to 90% (Rehman et al., 2009). Also, similar observations were reported previously by Rabbani et al. (2016). In conclusion, it is obvious that $Q_{Inlet\ air}$ and EBRT are crucial factors in BF design and performance. Due to the high sensitivity of this system, it needs more consideration to select these parameters in order to facilitate the reactor construction.

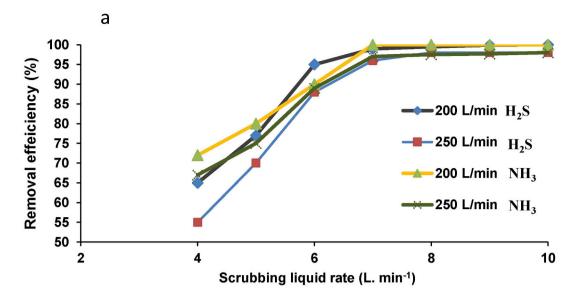
3.3. Chemical scrubber efficiency and removal rate study

3.3.1. Effect of gas flow rate and EBRTs in simultaneous removal of H_2S and NH_3 in the presence of oxidants

The performance of the CS was evaluated for efficient simultaneous removal of malodorous compounds including H_2S and NH_3 in terms of gas flow rate (100, 150, 200 and $250\,L\,\text{min}^{-1}$), EBRTs (6.46, 4.3, 3.23 and 2.58 s) and scrubbing flow rate (4–10 $L\,\text{min}^{-1}$) in the presence of three oxidants (NaOCl, H_2O_2 and $KMnO_4$).

Adsorption process coupled with chemical oxidation is responsible for the removal of H₂S and NH₃ in the CS. Therefore, the inlet flow rate, gas contact time and scrubbing liquid are crucial factors affecting H2S and NH₃ removal efficiency via mass transfer from the gas phase to the liquid phase. In this regard, the effect of air flow rate (100, 150, 200 and 250 L min -1 which considered areactor bed volume of $10.77 \, \text{L} \, \text{min}^{-1}$) and gas contact time (6.46, 4.3, 3.23 and 2.58 s) on H₂S and NH₃ removal efficiency was investigated in the presence of NaOCl, KMnO₄ and H₂O₂ oxidants as scrubbing liquid and depicted in Fig. 5. As can be seen in Fig. 5a, with increasing the air flow rate from $Q_{air} = 150-200 \,L\,min^{-1}$ and subsequently, decreasing bed contact time from 4.3 to 2.58 s, the H₂S removal efficiency decreases from 100 to 95% for NaOCl and KMnO₄ and also drops from 100 to 45% for H₂O₂. According to Fig. 5b, NH₃ removal efficiency does not vary too much because of low NH3 concentrations in the inlet gas to the CS. The removal efficiencies of 100, 98.5 and 96.3% by employing NaOCl and 100, 97.5 and 95% using KMnO₄ were obtained in the gas contact times of 4.3, 3.23, and 2.58 s, respectively. Statistic appraisal of the results, presented in Fig. 5, indicates that increasing superficial gas velocity (airflow rate) and subsequent reduction of gas contact time; the removal efficiency lessens regardless of oxidants used.

Increasing $Q_{Inlet \ air}$ up to $250 \ L \ min^{-1}$ makes an increase in pressure drop (ΔP) from 4 to 7.1 cm H₂O in the bed (Fig. 6), and hinders the mass transfer from gas to liquid phase ratio (L/G). So that, with decreasing liquid phase ratio, the removal efficiency declines too (Chen et al., 2001). Moreover, pressure drop which plays a key role in vertical upward flows is affected significantly by superficial gas velocity (U_{SG}). Accordingly, when U_{SG} increases, the scrubbing liquid acts as a hamper to the gas circulation from the bottom to the top of the contactor by forming a thick layer. Therefore, water drops are scattered into small drops, which are driven by the gas flow, a thin layer can be observed along the pipe walls and whereupon, the CS performance drops (Sanchez et al., 2007). Moreover, odor compounds are less water-soluble and then, with decreasing the gas contact time to 2.58 s, the H₂S and NH3 molecules could not completely transfer to the liquid phase and caused a negative impact on the CS removal efficiency (Albright, 2009; Moosavi et al., 2005). The high performance of CS for all oxidants in EBRTs of 4.3 and 3.23 s is attributed to the use of media with high specific surface area (plastic is one of the selective media in the CS because of less susceptibility to collapsing and clogging (Feilberg et al., 2010), hence, in this study PE was used with a high surface area), optimum rate of scrubbing liquid flow and mainly long retention time of



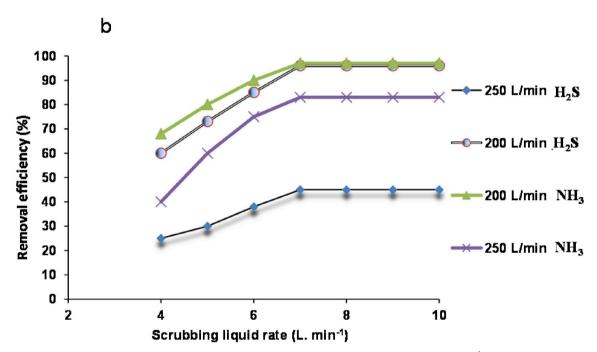


Fig. 7. Effect of scrubbing liquid on CS performance in removal of H_2S & NH_3 in airflow rates of 200 and 250 L min⁻¹. (a) NaOCl and KMnO₄, (b) H_2O_2 .

Table 4 Initial investment cost in systems, (a): BF and (b): CS.

Initial investment cost parameters	Cost (€)	
a		
Design and construction	133	
Packing material	76	
Supportive equipment costs (diffusing pump)	21	
Total initial investment cost	230	
b		
Design and construction	120	
media	12	
Humidifier & other equipment	47.5	
Total initial investment cost	179.5	

Table 5Initial operating cost in systems, (a): BF and (b): CS.

Initial operating cost parameters	Cost (€.year ⁻¹)	
a		
Electricity	210	
Water consumption	0.0744	
Labor	4380	
Total yearly operating cost	4590.0744	
b		
Electricity	70.5	
Water consumption	122	
Labor	4380	
Chemical usage	12768	
Total yearly operating cost	17340.5	

Table 6 Total annualized cost and annualized cost parameters for each system, (a): BF with airflow rate of $36 \, \mathrm{L\,min}^{-1}$ and EBRT 15 s; (b): CS with air flow rate of $250 \, \mathrm{L\,min}^{-1}$ and EBRT 2.58 s.

Annualized parameters	Cost (€.year ⁻¹)	
a		
AIC	40.244	
AOC	29835.48	
APMRC	32.48	
Total annualized cost	29908.2	
b		
AIC (€/year)	51.566	
AOC (€/year)	112713.5	
APMRC (€/year)	18.56	
Total annualized cost (€/year)	112783.62	

gas in the CS. These claims have been confirmed by previous studies (Charron et al., 2004; Chen et al., 2001). As can be seen from Fig. 6 (a & b), H₂O₂ has a low removal efficiency among other oxidants in EBRT = $2.58 \, \text{s}$ for H_2S (45%) and NH_3 (89%). The oxidative properties of H₂O₂ have a direct bond with the pH of the scrubbing liquid and its high performances have been observed in alkaline PH_S. With increasing the pH in the scrubbing liquid, the concentration of HS increases corresponding with improved alkalinity in the oxidant liquid. Thus, this causes not only the enhancement of the H2S solubility but also increases mass transfer between the gas and water phases (Moussavi et al., 2008). As a result, the pH range of 7.8-8.1 in the scrubbing solution according to Table 3 is not favorable to have high removal efficiency as a result of using H₂O₂. These results are consistent with previous findings (Moussavi et al., 2008). Also, similar observations have already been reported in various studies (González-Sánchez et al., 2008; Jiang et al., 2009; Karageorgos et al., 2010). This study was accomplished without the addition of any pH adjusters into the scrubbing liquid because one of the aims of this study was that all comparative data of two systems are collected in real circumstances in the field. The results indicated that EBRT associated with inlet airflow rate (Q_{Inlet air}) and type of oxidant are the main parameters in reactor design and its performance from a technical and economic point of view.

3.3.2. Effect of scrubbing liquid rate on reactor performance

Adsorption is directly proportional to the content of the liquid exposed to the gas stream. Therefore, the scrubbing liquid rate is a critical factor to achieve an optimum operation in the CS designing.

In this regard, the effect of scrubbing liquid flow rate in the range of $Q_L=4-10\,L\,\text{min}^{-1}$ was surveyed and presented in Fig. 7.

According to Fig. 7a, increasing the liquid flow rate from 4 to 7 $L.min^{-1}$ in $Q_{air}=200$ and $250\,L\,min^{-1}$ raises the H_2S and NH_3 removal efficiency from 65% to 55% to above 95% for NaClO and KMnO4 oxidants, respectively. A similar trend in the removal efficiency was also observed with the increase of the H_2O_2 flow rate, as shown in Fig. 7b. The removal efficiencies culminate from low ranges to above 40% and above 70% in $Q_{Inlet\ air}=250\,L\,min^{-1}$ for H_2S and NH_3 , respectively. Thus, the findings indicate that the changes in Q_L had a direct effect on the CS performance in the simultaneous removal of H_2S and NH_3 gases. Considering that Q_L and superficial liquid velocity (U_{SL}) are directly related, the U_{SL} affects the removal process in two ways. First, with increasing U_{SL} , mass transfer parameters enhance due to the supported

the diving force. Second, the scrubbing solution in high U_{SL} is regenerated, which under these conditions oxidant concentration is high and reaction velocity improved consequently.

Furthermore, for avoiding dry spots, which could be a bypass for untreated air leaving the CS, the scrubbing solution should be uniformly distributed at the top of the packing material (Sanchez et al., 2007). Then, in Q_L below $7 \, L \, min^{-1}$, these dry areas decrease the CS performance against gases treatment. In contrast, in $Q = 7 \, Lmin^{-1}$ and, above the CS bed has overcome the gas to liquid mass transfer limitation, which is wetting media ratio and reaches to high removal efficiency. According to the results presented in Fig. 7, it is obvious that the scrubbing flow rate = of $7 \, L \, min^{-1}$ is suitable Q_L for achieving high performance in the CS and is in good agreement with that reported in the literature (Moussavi et al., 2008).

3.4. Economic analysis

3.4.1. Investment cost

The cost analysis for both systems (CS and BF) at Yazd WWTP was divided into three chief parts: initial investment cost, operating cost, and replacement packing material cost. All these costs and their details are presented in Table 4 (a & b). Investment cost mainly depends on the volume of reactors, which is determined by various parameters such as the system's EBRT, airflow rate, concentration and type of the pollutant, and required removal efficiency. In this study, investment cost was classified into the three main parts; design & construction, media, and other supportive equipment (water bath in the BF and diffusing pump in the CS and so on) cost. Design & construction cost was estimated 120 and 133 € for the BF and CS, respectively. Forasmuch as volumes, and design factors were approximately close to each other and similar material was used to construct both systems, there was no significant difference in this part of the costs. Considering that the main portion of the BF bed is made using compost, which was cheaper than PE material (CS packing material), the cost of this section was lower in the BF system. Other supportive equipment costs were the last factor for estimated total investment cost in systems (47.5 € for BF and 21 € for CS). In this area, piping cost and other details cost were similar in two methods, but the water bath in the BF versus a diffusing pump in the CS made a difference in this section of fees. Plexiglas, which was applied to the constructed water bath in the BF, was worthy rather than diffusing pump in the CS. All in all, the design and construction sector was a highly expensive one above investment cost, and the total initial investment cost was 179.5 € and 230 € for the BF and CS, respectively. As a result, this part of the cost can be very different study-by-study because it mostly it depends on process design parameters, commodity prices, time, and location of study.

3.4.2. Operating cost

Operating cost is expressed as the yearly cost in Euros (ε .year⁻¹). In the CS system operating costs include the electricity, water consumption, labor, and chemical usage costs, while the BF system also includes all of these costs except chemicals. The BF worked more times than the CS and, its electricity usage cost (210ε .year⁻¹) was more than its opponent (70.5ε .year⁻¹). Moreover, the water consumption costs were 0.0744ε .year⁻¹ and 122ε .year⁻¹ for the BF and CS, separately. In fact, the CS is a water-based system, hence, for better performance, the

Table 7Economic comparison of CS and BF systems in different literature.

System	EBRT (s)	Airflow rate (m ³ /h)	Annualized cost (€/year)	Index (€/m³.h)	Reference
BF	60	20000	42000	1.8	Prado et al. (2009)
BF	60	50000	-	2	Estrada et al. (2012)
CS	70	17000	14000-23000	-	(Gabriel and Deshusses, 2004)
CS	4	50000	-	3.6	Estrada et al. (2010)

whole surface of media needs to be covered with water. Also, chemical usage is a highly expensive sector in the CS and affects total operating costs in this system. Because of the high fraction of NaOCl consumed $mg/(H_2S + NH_3)$ mg 6.5–7.1 and the liquid quantity of flow rate $(7 \, \text{L} \, \text{min}^{-1})$, it was an expensive one of the operating costs. Labor cost $(4380 \, \text{C.year}^{-1})$ was the same in both systems because the work cost unit was equal. In summary, chemical usage in the CS and work cost in the BF were identified as a costly part in reactors, and total initial operating costs were 4590.0744 C.year^{-1} and 17340.5 C.year^{-1} for the BF and CS, respectively. In conclusion, operating costs are an expensive segment in both systems costs Table 5 (a & b). This claim has been confirmed by previous studies (Estrada et al., 2012; Gao et al., 2001).

3.4.3. Packing material replacement cost

Packing material replacement cost considers a price to remove old bed materials, purchasing and installing the new one into the systems. The durability of the bed for the CS and BF was estimated 10 and 2 years, respectively. Based on the findings, the packing material replacement costs were 25 ϵ and 89 ϵ for the BF and CS systems, respectively. Considering the above, the main reason for the difference between costs in this section can be due to packing bed components.

3.4.4. Determining annualized costs in systems

The annualized worth (AW) method is customarily utilized for comparing alternatives. In this context, we estimated all annualized costs for all price section of rectors and finding shown in Table 6 (a & b). Total annualized cost for CS in airflow rate of $250 \, \mathrm{L} \, \mathrm{min}^{-1}$ and EBRT 2.58 s is 112783.62 €.year $^{-1}$, and for the BF it is 29980.2 €.year $^{-1}$ in airflow rate of 36 L min⁻¹ and EBRT 15 s. According to the results, the annual costs related to the CS are dramatically higher than BF. Regarding the volume of treated waste air by CS during a year and some operating costs (chemical usage), despite the shorter working time of the CS, this phenomenon is justifiable. The $(\in .m^{-3}. h^{-1})$ was applied as an index for comparing two alternative systems with the economic aspect for removal of odor from WWTP. This index was obtained 1.58 €.m⁻³. h⁻¹ for the BF with 18921 m³ year⁻¹ volume of treated air in EBRT 15 s and 2.57 \cdot .m⁻³. h⁻¹ for the CS with 43800 m³ year⁻¹ volume of treated air in EBRT 2.85 s. Table 7 indicates the result of various researches for economic analysis of the BF and the CS in different conditions.

4. Conclusions

The techno-economic comparison of the CS and BF systems employed for treatment of the real waste air streams containing NH3 and H₂S was conducted over a 45-day time period at Yazd wastewater treatment plant. The experimental results showed that the removal efficiencies of NH3 and H2S in both systems were affected by their respective loading rate. Additionally, the NaOCl solution, especially with the optimum flow rate of 7 L min⁻¹ was found to give the best results in term of removal efficiency among the various scrubbing liquids (NaOCl, H₂O₂, and KMnO₄) examined in the CS. The technical analysis clearly demonstrated the effectiveness of both systems in treating actual waste gases containing H2S and NH3. The CS is able to simultaneously treat of H₂S and NH₃ with efficiency above 97% for each compound at $Q_{air} = 250 \, L \, min^{-1}$ in the presence of NaOCl as an oxidant. However, the BF had the robustness to remove a yield above 92% and 99.5% for H_2S and NH_3 , respectively, at $Q_{air} = 36 \, L \, min^{-1}$ under suitable operating circumstances. Regarding the volume of treated contaminated air, it is obvious that the performance of the CS is better than BF. The economic index (ℓ .m⁻³. h⁻¹) was calculated by the annualized worth method and utilized the economic assessment of both control systems. The economic index for the CS and BF were estimated to be 2.57 and 1.58 €.m⁻³. h⁻¹, respectively, indicating the cost-effectiveness of the bio-filtration system compared to the CS ones. The results also revealed that chemical usage and operating costs were expensive parts of the CS

and BF, respectively. Although the BF system is preferred from the economic viewpoint, it suffers from some of the operational drawbacks such as performance fluctuation, low biodegradation rate, maintenance of biomass, and disinfection adequacy of the BF effluent which hinders the process from being widely employed and established. Accordingly and regarding technical results, the CS seems to be more efficient applicable odor control technology in treating off-gas of Yazd WWTP.

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Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.jenvman.2019.04.003.

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