



Pulp and paper mill effluent treatment by hybrid anaerobic upflow fixed-bed bioreactor combined with slow sand filter

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ABSTRACT

In the present work, pollutant removal study of Century Pulp and Paper Mill effluent was done. A hybrid unit of upflow fixed-bed anaerobic bioreactor (UFBAB) along with slow sand filter (SSF) was compared with single unit UFBAB for pollutant removal at different hydraulic retention time (HRT). The hybrid system showed better removal efficiency which can be attributed to SSF. It was observed that SSF provides a polishing effect to the effluent treated by UFBAB. The biodegradation rate in both filters was observed to be increased in first six weeks of experiment, prior to the acclimatization of microbial communities. The relative removal efficiency of biological oxygen demand and chemical oxygen demand was found to be between 0.90 and 0.99, respectively, at 24 h HRT, demonstrating very high removal efficiency of the hybrid system. The relative removal efficiency of total dissolved solids and total suspended solids was around 1 at 16 h HRT indicating 100% efficient and within the limits as prescribed by Central Pollution Control Board India (CPCB) for treated effluent discharge in surface water bodies. Best removal efficiency was observed in case of sulfate at low HRT of 8 h. Relative removal efficiency for phenols was found to be greater than 1 at 12 h HRT. SSF was found to be a suitable substrate for polishing of treated effluent of pulp and paper mill wastewater, demonstrating promising relative efficiency.

Keywords: Anaerobic biofilter; Slow sand filter; Polishing effect; Pulp and paper mill effluent

1. Introduction

The problem of water pollution is one such havoc that has taken the attention of scientists and social activists all round the globe. The main problem with wastewater is its generation in larger quantity with higher organic loads and serious pollution potentials

[1]. Therefore, problem of wastewater disposal continues to be a serious threat in present as well as in nearby future. Moreover, the growth of industrialization has led to search for disposal options that are environmentally safe and economically viable [2,3].

Paper industry consumes about 250–300 m³ water per tones of paper produced and generate an equal amount as wastewater [4]. In India, around 75% of total

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fresh water supplied to pulp and paper industries emerges as wastewater which requires an effective method of treatment [5]. The wastewater from pulp and paper industry mainly consists of chlorinated lignosulfonic acids, chlorinated resin acids, chlorinated phenols, and chlorinated hydrocarbon [6]. The important pollutants of this industry include color that is commonly visible over long distances in receiving rivers and streams. The color present in the waste is due to three-dimensional heterogeneous polymers composed of oxyphenylpropane units known as lignin and its derivatives. Lignin and its derivatives react with the chlorine content (discharged from bleached plant) those results in high biological and chemical oxygen demand (COD) of discharged wastewater [7]. Other major contaminants formed in the effluent of pulp and paper mill are solid residues such as sludge and absorbable organic halides (AOX) which are organically bound chloride, bromide, or iodide in either dissolved or suspended form [8,9]. Lignin, vanillin, vanillic acid, dehydrodivanillin, ferulic acid, cinnamic acid, syringic, catechol, coniferyl, alcohol dehydrodihydroconiferyl alcohol, phenyl propionic acid, and chlorophenols are the major by-products of the pulp and paper mill. In 1996, 76% of phenols released in aquatic bodies of Canada were released from the pulp, paper, and wood industry. The phenolic waste from pulp and paper industry may contain cyanide, aldehydes, ketones, alcohols, organic acids, and gases [10]. Photooxidation, oxidation, and microbial degradation are expected to be the major biotic process for the removal of phenol from the polluted waste stream [11]. Phenol that is the chief pollutant of paper and pulp industry, serves as a carbon source and hence can be easily utilized by the number of isolated and adapted microorganisms.

Release of pulp and paper mill effluent into natural waterways damages the water quality and causes a variety of clastogenic, carcinogenic, endocrine, and mutagenic effects on aquatic organisms [12]. Some compounds in the effluents are resistant to biodegradation and can bioaccumulate in the aquatic food chain [13,14]. The high polluting potential of pulp and paper industry wastewater can no longer be ignored and a need of efficient and economical treatment of these effluents still remains elusive [15]. The conventional pulp and paper mill wastewater treatment mainly use biological methods which involve aerobic/anaerobic microorganisms [16]. Bacterial colonies used in secondary wastewater treatment remove small organic carbon molecules by consuming them as a source of organic carbon. This results in growth of microbial mass and cleaning of water at the same time. Up to 80–90% of biological oxygen demand (BOD) is reduced during secondary treatment [17].

Anaerobic fixed-bed is an energy efficient process used for the treatment of waste containing high concentration of biodegradable organic material. It involves a series of degradation and fermentation reactions carried out by various prokaryotes to digest suspended solids and large macromolecules into soluble components. These products are further fermented by the cooperative actions of syntrophic bacteria to produce acetate, CO₂, and H₂ and finally broken down by methanogenic Archaea to produce methane and CO₂ [18].

Slow sand filter (SSF) is one of the most efficient processes improving the physical, biological, and chemical quality of water [19,20]. It relies on both physical and as well biological approach for waste treatment. Wastewater particles larger than the sand particles are easily strained out and retained within the filter making it one of the most efficient processes for improving the physical, biological, and chemical quality of wastewater. The most striking advantages of SSF involve its reuse, low energy requirement, tolerance of seasonal input fluxes and do not require lengthy start-up and shut down periods [21].

Keeping in view the benefits of microbial biodegradation SSF was used in combination with upflow fixed-bed anaerobic bioreactor for the treatment of pulp and paper mill effluent. From economical and operational point of view, biological treatment is energy efficient way of treating biodegradable wastewater [22] and this in combination with SSF with effective size 0.15–0.3 mm provide void passage for treated water that increases removal efficiency range by 99–99.9% [23].

2. Material and methods

2.1. Effluent collection

The raw effluent sample was collected from Century Pulp and Paper Mill, Lalkuan, district Nainital, India. The samples were collected by grab method from effluent discharge site and were transported within 4 h under refrigerated condition to laboratory and analyzed as per given in standard methods for examination of water and wastewater (APHA) [24].

2.2. Bioreactor design

In order to study the pollutant removal efficiency at different hydraulic retention time (HRT) two bioreactors, A (hybrid anaerobic upflow fixed-bed bioreactor combined with slow sand filters in series) and B (anaerobic upflow fixed-bed bioreactor) were designed (Fig. 1). These biofilters were set in duplication as A1,

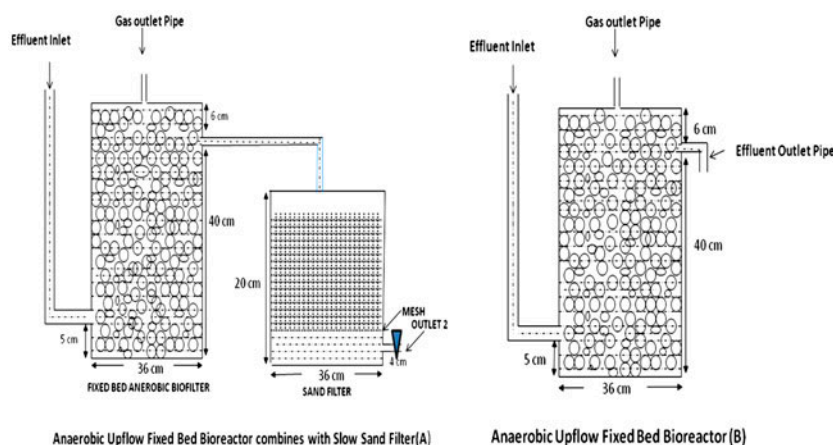


Fig. 1. Biofilter designed for pulp and paper mill effluent treatment.

A2 and B1, B2 to standardize the results obtained. The bioreactor had an internal diameter of 36 cm, height 45 cm, and volume of 45 L. The sand filter attached with bioreactor A1 and A2 had an additional volume of 20 L. The anaerobic biofilters had an inlet 5 cm and an outlet at the height of 40 cm from the base. A gas outlet at the top of upflow fixed-bed bioreactor was also provided for the release of gases produced during anaerobic treatment. Both the anaerobic upflow fixed-bed bioreactor consists of gravel (2.5–3.8 cm diameter) as support material. Gravels act as support for naturally self-immobilization slime layer formed by anaerobic microbes. Immobilization of microbial cells has received increasing interest in the field of waste treatment and offers advantages such as high biomass, high metabolic activity, and strong resistance to toxic chemicals [25]. Moreover, immobilized microorganisms are cost-effective and can be used several times without significant loss of activity in anaerobic bioreactors. Effluent moved in a vertical upflow mode, thus got maximum contact time with slime and got treated while moving upward in the bioreactor. The sand filter attached to bioreactors A1 and A2 had sand particles between the range of 1.7 and 2 mm. This sand passed sieve opening 2 mm and was retained on sieve opening 1.7 mm, hence eliminating particles above and below this range. The selected sand particles were then washed to remove the clay and organic matter adhering to them.

2.3. Acclimatization of bioreactor

Microbial species adapt themselves to new environmental factors over time by a phenomenon known as acclimatization. During acclimatization, biodegradation rates are influenced by the duration of acclimatization and the degree of microbial toxicity imposed by

the new environmental factors [26]. Prior to effluent treatment, all the bioreactors were kept for 6 weeks acclimatization period during which it was supposed that the active growth of bacteria is leading to slime formation around the gravel surface. The effluent to be fed in the bioreactor for acclimatization was especially prepared. One kilogram of fresh settled sludge (source of microbial inoculum) from the effluent treatment plant of pulp and paper mill, 1 L of cow dung slurry (for inoculation of methanogenic microorganism), 4.5 gm of KMnO_4 to fix free oxygen, and 200 gm of nutrient broth media (to provide initial nutrient source for actively growing bacteria) were mixed with 10 L of effluent and were filtered with 2 mm sieve. This solution was placed in well sealed fixed-bed upflow bioreactors at HRT of 3 d. At the interval of 3 d, 20% of the acclimatizing feed was drained out of the biofilter and replaced by pulp and paper mill wastewater. In biofilters A1 and A2, this draining out of effluent was done through additionally attached sand filters, so that sand filter also gets inoculated for future polishing of treated effluent. The replacement of drained feed by fresh effluent was repeated periodically at the interval of 3 d till 6 weeks of acclimatization.

2.4. Bioreactor start-up

After the period of 6 weeks of acclimatization period, it was supposed that the active growth of bacteria has led sufficient slime cover around the packing media in all the bioreactors, i.e. A1, A2 and B1, B2. Before actual startup of bioreactor, new effluent is fed in bioreactors replacing continuously the effluent at a HRT of 16 h to give a good reactor startup and adapt the microorganism for new feed, for continuous and

efficient operation. After a week of effluent flow in the bioreactors, the results were analyzed at different HRT of 8, 12, 16, 20, and 24 h. The HRT was calculated as follows:

$$\text{HRT} = \frac{\text{Volume of biofilter}}{\text{Effluent flow rate}}$$

SI unit volume is in (m³) and effluent flow rate is in (m³/h). HRT is usually expressed in hours or days. Hence, HRT was maintained by adjusting the flow rate of the effluent, such that it spends the desired time inside the bioreactor. The reduction was measured by analyzing different physico-chemical parameter in triplicate from each biofilters. The mean value of both the replicate biofilter setup (A1, A2 and B1, B2) was taken for further calculations.

2.5. Analytical methods

The effluent was characterized for various physico-chemical parameters like pH, COD, BOD, total dissolved solids (TDS), total suspended solids (TSS), sulfates, chlorides, and phenols, and each parameter was analyzed in triplicate in order to see the standard error in handling. All analyses were carried out as per standard procedures given by APHA [27]. The biofilter performance was characterized by relating influent water quality to effluent water quality to calculate efficiency ratio and relative efficiency. Efficiency ratio was calculated as, ER = 100 – (average outlet/average inlet) * 100. Relative efficiency involves a base concentration level (CPCB limit of effluent release in surface water bodies) to relate to influent and effluent concentrations, rather than removal based on the change of concentration from influent to effluent. The equation, relative efficiency = (average influent pollutant concentration – average effluent pollutant concentration) / (average influent pollutant concentration – CPCB

water quality standard for discharge), presents the generalized relative efficiency formula [28]. Studies were conducted to determine the pollutant removal and polishing effect provided by SSFs under different HRT of 8, 12, 16, 20, and 24 h.

3. Results

3.1. Characterization of pulp and paper mill effluent

The Century Pulp and Paper Mill effluent was found to be alkaline in nature with pH value ranging from 8.6 to 9.2. Apart from high COD and BOD, the pulp and paper mill effluent also contained high dissolve and suspended solids, the concentration of sulfates, chlorides, total nitrogen, and phenol concentration values were quite higher than permissible limits as prescribed by CPCB for discharge of wastewater in inland surface water [29]. The pulp and paper mill effluent load and the CPCB prescribed limits of inland wastewater discharge are displayed in Table 1. Analysis of the data demonstrated that the characteristics of the pulp and paper mill effluent were in agreement with the reported values by other researchers [30,31]. However, variation in some parameters can be attributed to the raw material, chemicals, and methodology adopted during wood pulping and paper making by respective mill.

3.2. Performance of biofilters at different HRT

Sand filters have been mostly used as a cost-effective alternative to conventional septic tank/soil adsorption systems for domestic wastewater [32,33]. However, their use for treatment of industrial wastewater with high pollution load still remains untested. In this laboratory study, the best performance is achieved when a hybrid upflow fixed-bed bioreactor (A) is attached to sand filter, Table 2 proves this by comparing relative efficiency (A) and (B) bioreactor.

Table 1
Wastewater characteristics of the Century Pulp and Paper Mill effluent

Century Pulp and Paper Mill effluent characteristics	Effluent value (mg/L)	CPCB general standards for discharge of environmental pollutants (mg/L)
pH	8.9	5.5–9.0
Biological oxygen demand	1,280	30
Chemical oxygen demand	2,912	250
Total dissolved solid	1,270	500
Total suspended Solid	1,265	100
Sulfate	207	200
Chloride	365	250
Phenols	2.3	1.0

Table 2

Relative efficiency of upflow anaerobic fixed-bed bioreactor with SSF (A) and control upflow anaerobic fixed-bed bioreactor (B)

HRT	8		12		16		20		24	
	A	B	A	B	A	B	A	B	A	B
BOD	0.53	0.26	0.64	0.47	0.77	0.60	0.87	0.63	0.90	0.72
COD	0.78	0.42	0.92	0.59	0.96	0.74	0.97	0.76	0.98	0.80
TDS	0.79	0.41	0.83	0.60	0.90	0.67	1.01	0.79	1.03	0.88
TSS	0.99	0.54	1.06	0.65	1.1	0.89	1.11	0.90	1.13	0.93
Sulfate	1.04	0.15	1.43	0.42	1.75	0.64	1.85	1.02	1.95	1.45
Chloride	0.78	0.17	0.95	0.56	1.24	0.66	1.54	0.87	1.64	1.05
Phenols	0.74	0.27	1.03	0.45	1.14	0.65	1.25	0.73	1.34	0.82

3.2.1. pH adjustment

The pH of the effluent which was initially 8.9 attained a constant pH of 7.4–7.2 at a HRT of 16 h in biofilter hybrid A, whereas in biofilter B, the constant pH attainment occurred at 20 h depicting that in hybrid biofilter (A), the methanogenic phase could start efficiently at much lower time compared to biofilter B as methanogens need a near neutral pH to work and as soon this pH is attained, the degradation work start [34].

3.2.2. BOD removal

At HRT of 8 h, the BOD removal in biofilters A and B shows a pollutant removal efficiency of 49.2 and 23.7%, respectively (Fig. 2). With the increase in HRT of 12 h, performance ratio also increases linearly to 59.3 (A) and 43.4% (B), this linear increment in performance was seen up to the HRT of 16 h in both the biofilter and attain a pollutant removal efficiency of 71.6 and 55.6%, respectively, although there was increment in performance up to 81 and 83.5% for biofilter A, and 58.2 and 66.8% for biofilter B at 20 and 24 h HRT, but with respect to time, the slope as shown graphically (Fig. 2) is nearly parallel to x -axis symbolizing stationary phase or the maximum removal capacity of biofilters, with the combination of organic matter and microbial culture present in effluent. It is suggested that the change in slope of curve may have been due to the polishing effect provided by SSF. According to Uyanik et al. [35], the change in the slope of curve after 16 h HRT may be due to strong wastewater containing high organic load. On partial degradation of these substrates, significant amounts of fatty acids are formed, which enhanced the biological oxidation up to a certain point. After this point, degradation rate and methanogenic population in the reactor are inhibited. The pollutant removal was also studied as relative reduction efficiency of pollutant in

respect to discharge limits of CPCB. In biofilter A, it was 0.53, 0.64, 0.77, 0.87, and 0.90, and for biofilter B, the values were 0.26, 0.47, 0.60, 0.63, and 0.72 at 8, 12, 16, 20, and 24 h HRT.

3.2.3. COD removal

Similarly, in case of COD at HRT of 8 h, the biofilters showed pollutant removal efficiency of 70.2% for A and it was 37.7% for B; the difference of 1.2-fold in the values can be credited to SSF. This difference of treatment efficiency was also seen at HRT of 12 h 82.6% (A) and 52.7% (B), and 16 h 86.7% (A) and 66.3% (B) after which the pollutant removal efficiency increment does not take place with the same rate and a minor increment in the initial values was seen at HRT of 20 and 24 h, indicating the arrival of stationary phase or the maximum COD removal efficiency. Fang and Chui [36] reported that the COD removal efficiency in their upflow anaerobic sludge blanket reactor (UASB) was mainly dependent on the COD loading rate and HRT of the reactor operation. But in other study done by Rajakumar and Meenambal [37], different wastewaters of high strength like poultry slaughter house wastewater; hybrid reactor was effective and resulted in 80 and 86% of COD removal efficiency. These results were also found in accordance with the hybrid biofilter B used in present study. In terms of relative efficiency, COD removal was found up to 0.78, 0.92, 0.96, 0.97, 0.98 for biofilter A, and 0.42, 0.59, 0.74, 0.76, and 0.8 for B, respectively, at 8, 12, 16, 20, and 24 h HRT.

3.2.4. TDS and TSS removal

SSFs can be best use to control the dissolved and suspended solids of pulp and paper mill effluent. It was found that biofilter A has a relative efficiency of 51, 53, 58, 65, and 66.5% for TDS and 83.7, 89.44, 92.9, 93.8, and 95.2% for TSS removal at 8, 12, 16, 20, and

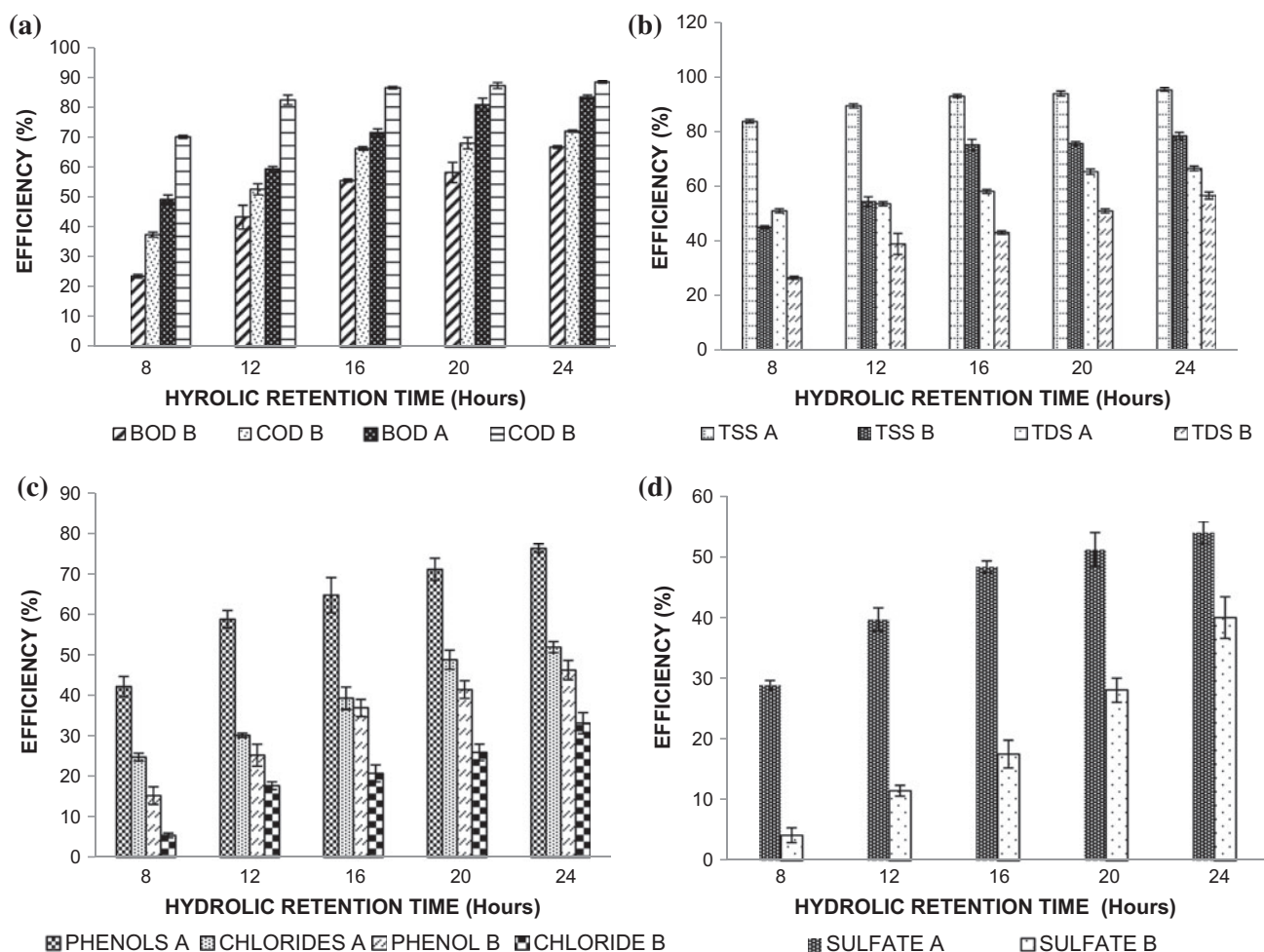


Fig. 2. Pollutant removal efficiency of biofilters A and B at different HRT. (a) BOD and COD removal efficiency of biofilters A and B at different HRT. (b) TSS and TDS removal efficiency of biofilters A and B at different HRT. (c) Chloride and phenol removal efficiency of biofilters A and B at different HRT. (d) Sulfate removal efficiency of biofilters A and B at different HRT.

24 h of HRT. The use of biofilter A was found to be 100% efficient at HRT of 20 h for TDS in a way that the treated effluent attained the effluent discharge limit of CPCB, whereas in case of TSS at 12 h of HRT, this limit was attained proving to be best for TSS and TDS control system. The TDS and TSS removal efficiency increased with the increase of HRT. These results were in accordance with Rolland et al. [38] experiment that to have a good removal of soluble organic matter, it is necessary to increase the residence time in the filter, particularly with the coarse sand.

3.2.5. Sulfate removal

At 8 h of HRT, sulfate shows a pollutant removal efficiency of 28.8 and 4.2% removal in biofilters A and

B that increased linearly till 16 h of HRT to attain value of 48.3, in biofilter A after 16 h, there was a negligible change in the pollutant removal efficiency. In biofilter B, this removal efficiency was seen to increase linearly with increase of HRT attaining value up to 40% efficiency at 24 h HRT. These findings similar to the study done by Bayrakdar et al. [39] who found out that sulfate-reduction efficiency quickly increased during the start-up period and reached 80% within 45 d, and the biofilm type reactors have higher removal rates at short HRT. Decrease in the performance at higher HRT may be due to reduction of sulfate into sulfite, and it is speculated that these reduced sulfite may had inhibitory effect in metabolism of methanogens. Many researchers have reported sulfide toxicity to methanogens, and microorganism become inactive and

reduces the conversion of intermediate to final end products leading to their accumulation and hence reduce efficiency [40,41].

3.2.6. Chloride and phenols removal

In chloride removal, the incorporation of SSFs provided a good relative efficiency of 24% in biofilter A against the biofilter B which was 5.4% at similar HRT of 8 h. The SSFs increased the chloride removal nearly by four fold. As HTR was increased, the reduction efficiency also increased linearly till 20 h HRT after which the slope starts to approach a stationary phase in biofilter A, whereas in biofilter B, this had a linear approach till 24 h HRT. At 16 h of HRT, biofilter A attained the CPCB limit of discharge with relative efficiency of 1.24, whereas a low relative efficiency of 0.56 was calculated in biofilter B at same time showing sand filters to be very efficient in chloride removal. Similar results were seen for phenols reduction biofilter A showed relative efficiency of 0.74, whereas biofilter B had relative efficiency of 0.27 at HRT of 8 h. The biofilter attains relative efficiency of 1.14 at 12 h HRT and 1.34 at 24 h HRT which coincide with the CPCB limits of discharge. These findings were in accordance with Ananyeva et al. [42] who reported complete biodegradation of phenol within 70 h and Fahmy et al. [43] also studied the degradation of 2,4,6-trichlorophenol, 2,4-dichlorophenol, and 4-chlorophenol under anaerobic conditions, using an adapted biofilm in fluidized sand bed fermenters with a minimum retention time of 11 h, chlorophenols were almost completely removed.

Ferguson and Dalentoft [44] also observed 40–65% AOX removal in anaerobic treatment of bleach plant effluent. Savant et al. [45] also reported that maximum dechlorination was found under anaerobic conditions. Paszczynski et al. [46] have also reported that most of the chlorinated phenols and other low molecular mass components of the effluent were removed during fungal treatment, and according to Hakulinen and Salkinoja-Salonen [47], the chlorophenolic compounds mineralized in the bioreactor to non-toxic end products, CO₂ and chloride ions. SSFs provided a substrate of growth for this microbial and fungal biomass, and hence, biofilter A showed an increase in efficiency.

Biotic (48%) and abiotic (52%) factors were proportionately involved in the removal of phenolics and sand provided a suitable substrate for the treatment of phenolic-laden waste [48]. Prior acclimation of microbial communities also increase the biodegradation rate of phenolic acids significantly and their findings were also showed that biotic removal of gallic acid and vanillin (phenols) was, respectively, 6% and 12%

higher in the microcosms containing acclimated sediment than in the control microcosms. Welz et al. [48] also demonstrated that a period of nine weeks was sufficient for the microbial population in a sand filter to acclimate biotic degradation rates when compared to a non-acclimated population. These results were also supported by Blum et al. [49] and Vaughan et al. [50] that low concentrations of phenolic acid mixtures stimulated the growth of phenolic acid-utilizing bacteria within the bulk soil and that competitive selection of these bacteria enhanced biodegradation of phenolic acids. Interactions of methanogens and denitrifiers were also investigated by Fang and Zhou [51] in a UASB treating phenol (200 mg/L) and m-cresol (100 mg/L) containing wastewater. In their findings, over 98% phenol and 60% m-cresol were degraded jointly by methanogens and denitrifiers with 1 d HRT, to this, the combination of SSFs can bring even better results at lower HRT. In their work, Welz et al. [48] concluded that abiotic removal (SSF) was related to the physical structure and the chemical composition of the substrate with clay, organic carbon, and metals being strongly associated with phenolic binding and/or chemical transformation reactions. And abiotic phenolic attachment sites were finite and biotic removal must also occur to ensure the longevity of sand filters [48].

4. Conclusion

The polishing effect of SSFs was assessed by comparing hybrid anaerobic upflow fixed-bed bioreactor combined with SSFs (A) in series and anaerobic upflow fixed-bed bioreactor (B). It was concluded that a hybrid fixed-film bioreactor with SSFs (A) had numerous advantages such as enhancing microbial cell stability, allowing continuous process operation, avoiding the biomass–liquid separation requirement, low energy consumption, low chemical consumption, less equipment required, and high operational simplicity. And also reduce the effluent treatment time (HRT) and space requirement. In order to reduce environmental pollutions through biodegradation of many harmful compounds and harness, the full potential of the biological treatment process, it can be one of the most promising areas of research. The application of SSFs technology attached in its preliminary stages, but the results seen so far are promising. Further research on biogas capture system, nature of sludge produced, methods for settled sludge separation are needed. The effect of structure and the size of packing material in biofilters can also be an important study in biofilters. The composition of the medium, its pH, and environmental conditions

considerably influence the adsorption of cells by changing their electrokinetic potential; therefore, the nutritional requirements of the microorganisms (active slime) can also be studied.

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