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Effect of process variables on the treatment of toxic wastewaters with anaerobic GAC reactors

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Abstract

The expanded-bed anaerobic GAC reactor, operating with GAC replacement, was demonstrated to effectively treat hazardous wastes. This study investigated the effects of GAC replacement rates, organic loading, and hydraulic retention time (HRT) on the treatment efficiency of an inhibitory wastewater. A synthetic toxic wastewater containing 5 g/l acetic acid, 3 g/l phenol and 0.9–1.8 g/l ortho-cresol was fed at a constant flow rate to three anaerobic GAC reactors, two of which were operated at an unexpanded empty-bed HRT of 1.0 d and the third at a HRT of 0.5 d. The reactors affected more that 97% COD removal throughout the study. Excellent treatment of the toxic waste was accomplished at GAC mean residence times as low as 8 d, and at COD loading rates of 35 kg/m^3-d. The reactors converted over 90% of the biodegradable influent COD to methane gas at high GAC residence time. The methane conversion efficiency dropped to about 64% in the reactors operated at a HRT of 1.0 d and 80% in the reactor operated at a HRT of 0.5 d at GAC residence times of 8 d. The GAC replacement rates were sufficient to overcome the toxicity of ortho-cresol. Organic loading rates and HRT appeared to adversely influence process performance, particularly when phenol was predominantly removed by biodegradation.

1. Introduction

The treatment of toxic organics in groundwater, landfill leachates, and various industrial wastewaters has elicited the concern of governments, and industries alike due to increased public awareness of their potential health hazards and stringent environmental regulations. Adsorption onto activated carbon has long been a

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recognized treatment technology. Aerobic biodegradation in activated carbon filters has been implicated in extending the service life of GAC adsorbers [1–4]. More recently, the amenability of toxic organic wastes to treatment in fluidized-bed anaerobic GAC reactors has been well documented in the literature [5–7]. The adsorptive capacity of GAC for organics and the excellent microbial attachment characteristics of the GAC medium [8–10] are the primary reasons for the success of anaerobic fluidized-bed GAC reactors in the processing of toxic organic wastes.

Nakhla et al. [11] reported that the replacement of a portion of the reactor medium with virgin GAC on a regular basis maintained the concentration of toxicants below inhibition levels and permitted long-term continuous treatment of hazardous wastes such as coal gasification wastewater. The GAC replacement rate is mostly dictated by the concentration of the adsorbable nonbiodegradable inhibitory compound in a wastewater and the hydraulic retention time (HRT). However, the replacement of exhausted medium with virgin GAC reduces the biological solids retention time (SRT) in the reactor and subsequently the removal rates of the biodegradable constituents of the waste. Therefore in this treatment technology, since adsorption and biodegradation are both effective in contaminant removal, a strong interrelation between the HRT, SRT, and GAC replacement rate exists. The objectives of this study were to investigate the effect of GAC mean residence time on the performance of anaerobic GAC reactors, evaluate the influence of the organic loading rate and the hydraulic retention time on process performance, and assess the impact of inhibition on the treatment efficiency of toxic wastewaters. To realize the goals of this study, three anaerobic GAC reactors treating a waste mixture comprising the anaerobically biodegradable nonadsorbable acetate, and the biodegradable adsorbable phenol were employed. To assess the impact of organic loading and HRT, one reactor was operated at double the biodegradable COD loading of the other two. Inhibition effects were evaluated by introducing the anaerobically nonbiodegradable adsorbable *o*-cresol [12] to the feed wastewater for two reactors.

2. Materials and methods

Three identical completely mixed expanded-bed anaerobic GAC reactors were employed for the purposes of this study. Two reactors were operated at a hydraulic retention time (HRT), based on the unexpanded bed volume, of 1.0 d with one treating a synthetic wastewater containing acetic acid and phenol (Reactor 1) while the second reactor was fed a solution of acetic acid, phenol, and *o*-cresol (Reactor 2). The third reactor (Reactor 3) was used to treat a synthetic wastewater containing acetic acid, phenol, and *o*-cresol at a reduced HRT of 0.5 d. The composition of the organic substrate solution for each of the three reactors is presented in Table 1. The employment of identical GAC replacement rate in Reactors 2 and 3 necessitates the application of equal mass loading rates of the adsorbable nonbiodegradable waste constituent to ensure fair assessment of the impact of the biodegradable organic loading on treatment efficiency.

Table 1
Composition of synthetic wastewater

Compound (mg/l)	Reactor 1	Reactor 2	Reactor 3
Acetic acid	10,000	10,000	10,000
Phenol	6000	6000	6000
o-Cresol	—	3540	1800
Sodium hydroxide	3750	3750	3750
Sodium carbonate	3625	3625	3625
Sodium sulfide	156	156	156

Inorganic salts are added for pH adjustment at 7 pH units.

The completely mixed anaerobic GAC reactor is presented in Fig. 1. The 11 l reactor constructed of 134.6 cm long, 0.63 cm thick, 11.4 cm OD plexiglass tube was surrounded by a plexiglass jacket for the passage of heated water to maintain the reactor temperature at 35 °C. An internal recycle stream at 8.0 l/min provided an initial bed expansion of 25% and achieved completely mixed flow regime in the reactor. Each of the three reactors was initially charged with 1.5 kg of 12 × 16 US Mesh Filtrasorb 400 GAC (Calgon Corp. Pittsburgh, PA, USA) resulting in a consolidated bed height of 38 cm. However, while operating at a GAC replacement rate of 75 g/d, problems with bed growth and fluidization necessitated the reduction of each of the three reactors media to 1200 g.

The flow of the synthetic wastewater (Table 1) was supplemented by an equal flow of an inorganic nutrient and vitamin mixture, whose composition was reported by Suidan et al. [5]. The total influent flow rate to Reactors 1, 2, and 3 were maintained at 3 and 6 l/d, respectively. The GAC replacement rates, investigated in this study were 25, 37.5, 50, 75, 100, and 150 g/d resulting in GAC man residence times of 60, 40, 30, 16, 12, and 8 d, respectively. Effluent samples collected from the reactors were analyzed for TSS following the procedures in Standard Methods [13]. The chemical oxygen demand of the filtered and unfiltered effluent was determined using the Hach COD Reactor Model 16,500 (Hach Company, P.O. Box 389, Loveland, CO 80539). Effluent fatty acids and phenolic compounds content were determined via gas chromatography on a Hewlett–Packard model 5730 A gas chromatograph equipped with a flame ionization detector. Glass columns packed with 0.3% carbowax with 0.19% phosphoric acid on 60/80 carbopack, and 10% SP-2100 on 100/120 supelcoport (Supelco Inc., Bellefonte, PA) were used for the volatile fatty acids and phenols, respectively. Detector and injection port temperatures were maintained at 200 °C, and the oven temperatures for fatty acids and phenolics were set at 120 °C and 130 °C, respectively. The effluent suspended carbohydrates were analyzed following the procedure of Kampmeir [14]. Gas samples from the reactors were analyzed for methane, carbon dioxide, nitrogen and oxygen using a Fisher model 1200 gas partitioner (Fisher Scientific Co., Chicago, IL, USA).

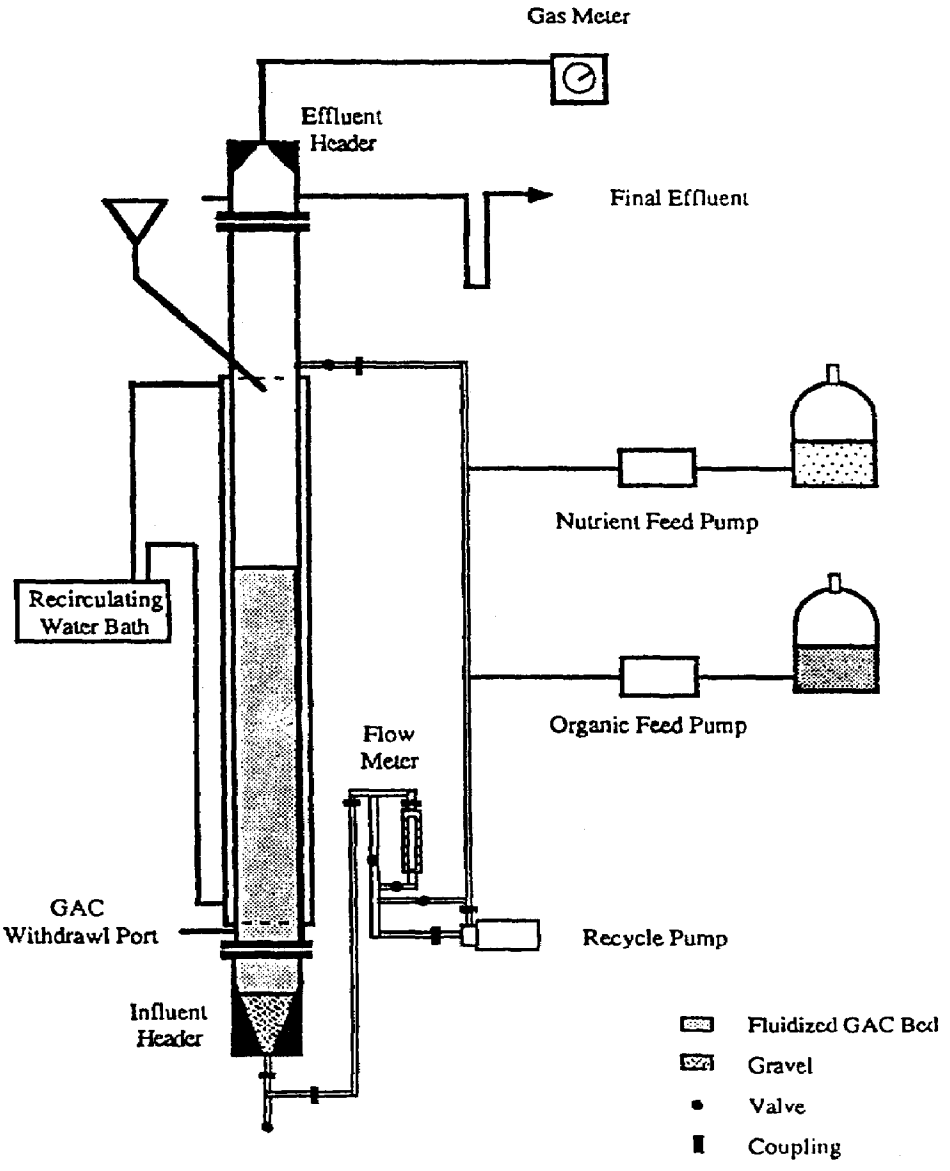

Fig. 1. Schematic of the fluidized-bed anaerobic GAC reactor.

3. Results and discussion

3.1. Removal of specific organics

Reactors 1 and 2 were initially operated at a biodegradable COD loading rate of 12.1 kg/m^3-d and subsequently increased to 15.1 kg COD/m^3-d upon reduction of bed GAC from 1500 to 1200 g while Reactor 3 was continuously operated at twice

that biodegradable COD loading rate. Reactors 2 and 3 were additionally fed cresol at an initial nonbiodegradable COD loading rate of 4.4 kg COD/m^3-d increasing to 5.5 kg COD/m^3-d after bed GAC reduction. At each GAC replacement rate, the reactors were operated to steady-state conditions, as indicated by the consistency of the effluent quality.

The temporal profile of the effluent acetate and phenol concentrations from Reactors 1, 2, and 3 are presented in Figs. 2(a)–(c) respectively. The numbers at the top represent the various daily GAC replacement rates which is also true of all figures to follow. Effluent acetate concentrations ranged from 15 to 100 mg/l in Reactors 1 and 2 and from about 25 to 110 mg/l in Reactor 3. Phenol data exhibited more scatter, however, varying from 0.1 to 7 mg/l in Reactor 1, 0.1 to 12 mg/l in Reactor 2, and 0.15 to 17 mg/l in Reactor 3. Thus throughout the study, the reactors removed over 99% of the influent phenol and over 97% of the influent acetate. In the anaerobic GAC reactor operating with GAC residence times, biodegradation and adsorption complement each other as pollutant removal mechanisms. At high GAC replacement rates, biodegradable adsorbable compounds such as phenol are utilized biologically, while at low GAC mean residence times corresponding to high GAC replacement rates adsorption becomes the dominant removal mechanism. Consequently while the methane productivity may deteriorate, the removal efficiency of phenol may not be significantly impacted.

A very interesting phenomenon was observed in the three reactors. The effluent acetate and phenol concentrations nearly followed the same trend. In Reactor 1, a significant drop in phenol and acetate occurred concurrently with the initiation of GAC replacement on 62 d. The decrease in effluent concentrations of biodegradable waste constituents with the reduction of biological SRT is inconsistent with substrate utilization kinetics. But as phenol is both adsorbable and biodegradable, the initiation of GAC replacement and the fresh adsorption capacity inducted in the system initially caused the rapid transient drop in effluent phenol. Furthermore since acetate has been shown to be an intermediate product of phenol biodegradation [15], the transient decrease of effluent acetate is concomitant with the drop in biological phenol utilization. Reactors 2 and 3 did not exhibit as much of a drop as Reactor 1 because the *o*-cresol present in the influent wastewater to Reactors 2 and 3, was adsorbed in preference to the phenol.

The effluent phenol and acetate concentrations from Reactors 1 and 2 increased continuously, following the initiation of the 100 g/d replacement rate, to reach their peaks and subsequently decreased until the end of the study. The simultaneous increase in the effluent concentrations of acetate and phenol, together with the drop in methane production observed during this period, indicates that biomass loss by shearing reduced the biological SRT significantly below the GAC mean residence time. At the 8 d GAC mean residence time, adsorption contributed appreciably to the removal of the biodegradable phenol, and since acetate is an intermediate of anaerobic phenol biodegradation [15], the drop in phenol concentration was therefore accompanied by a decrease in the effluent concentration of acetate. Both acetate and phenol effluent concentrations from Reactor 2 matched those from Reactor 1 for most of this study with the exception of the much sharper breakthrough of phenol at the

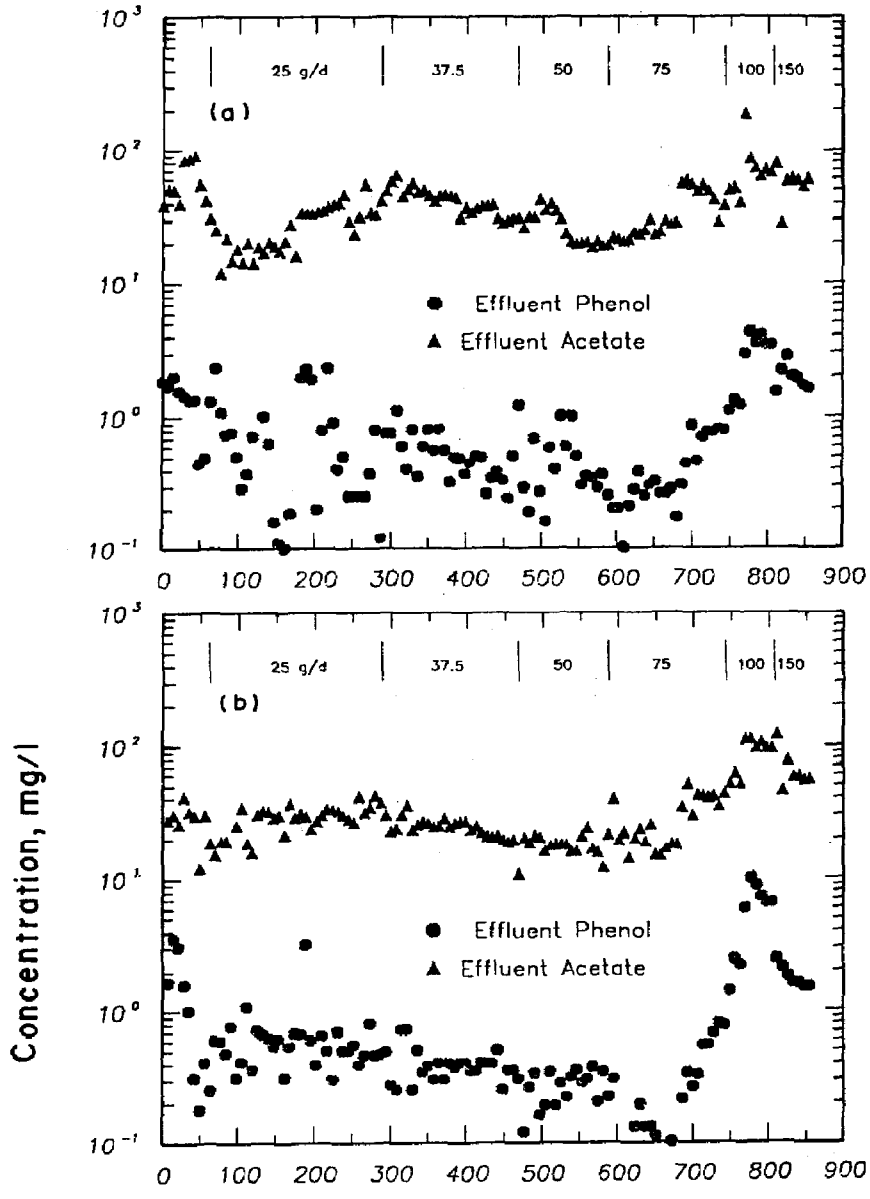

Fig. 2. Temporal profile of effluent acetate and phenol in Reactor 1 (a), Reactor 2 (b), and Reactor 3 (c).

12 d GAC mean residence time. Effluent concentrations of specific biodegradable organics were significantly influenced by the organic loading rate attested by the generally much higher effluent acetate and phenol data of Reactor 3 in comparison with Reactor 2.

Figs. 3(a) and (b) show the variations in effluent *o*-cresol concentrations from Reactors 2 and 3, respectively, with operating time. Following the initial increase in

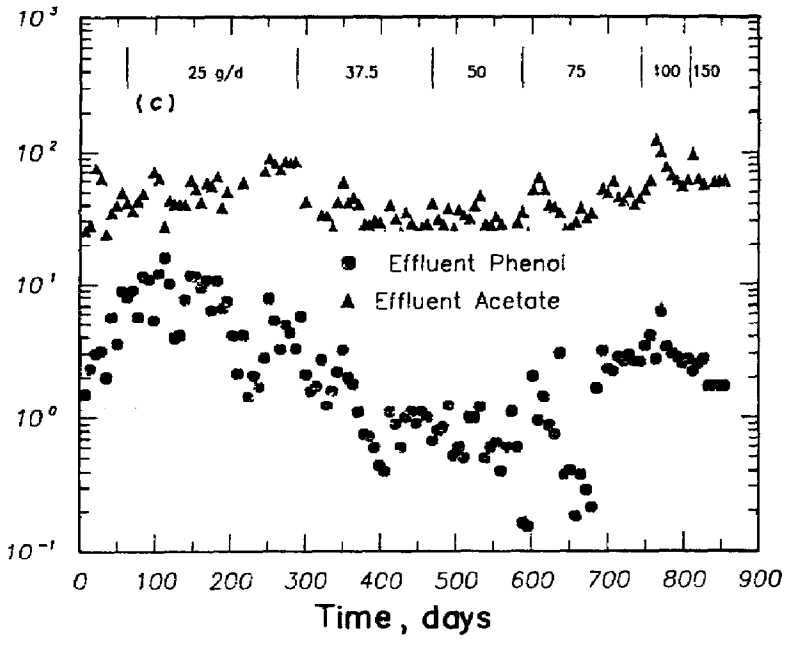

Fig. 2. Continued.

o-cresol concentration due to introduction into the feed wastewater, the effluent concentrations in both reactors decreased continuously throughout the study. This decrease was caused by the higher adsorptive capacities at increasing GAC replacement rates. Both reactors performed comparably with respect to cresol removal with the effluent concentrations ranging from about 50 mg/l to as low as 0.6 mg/l in both reactors. Since *o*-cresol was not biodegraded in the system [12, 16] and was only removed by adsorption, the application of identical GAC replacement rates and cresol mass loading rates ensured the attainment of similar effluent concentrations. Both reactors affected over 96% removal of *o*-cresol at the beginning of the study and as much as 99.99% at its end. Since phenol is less adsorbable on GAC than *o*-cresol at similar concentrations [17] and moreover effluent *o*-cresol was generally much higher than phenol, competitive adsorption effects on cresol adsorptivity were similar in both reactors despite the higher phenol concentrations detected in the effluent from Reactor 3.

3.2. COD removal and methane production

The temporal profiles of the influent, the total effluent, and the filtered effluent COD concentrations for the three reactors are presented in Figs. 4(a)–(c). All three reactors affected over 95% removal of the influent COD throughout the study. As depicted in Fig. 4, the three reactors exhibited the same trend of decreasing total effluent COD concentrations with increasing GAC replacement rates. As will be seen later, this decrease in the total effluent COD concentration was primarily caused by

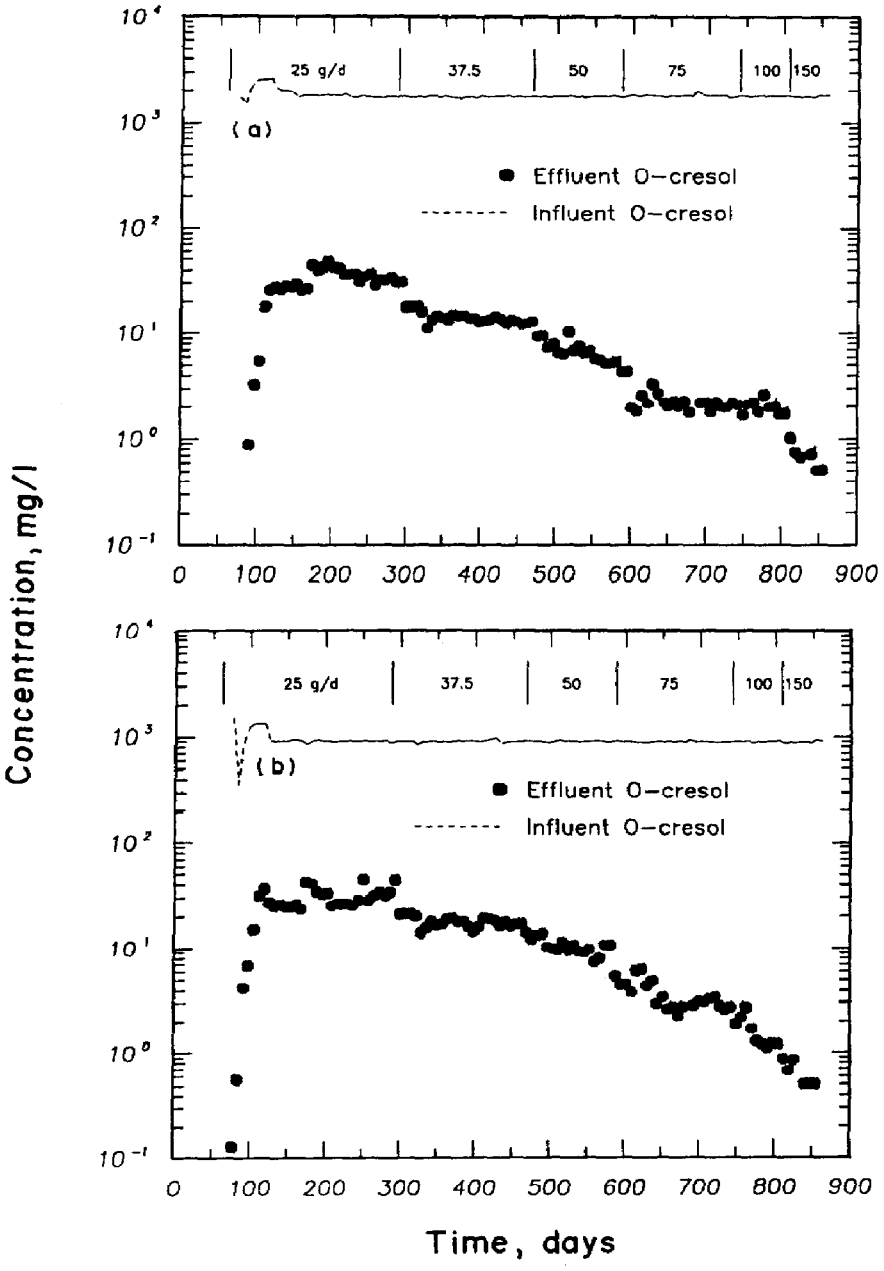

Fig. 3. Temporal profile of effluent *o*-cresol in Reactor 2 (a) and Reactor 3 (b).

a reduction in the concentration of suspended solids in the effluent. Fig. 4(a) shows that the filtered effluent COD concentration from Reactor 1 remained relatively constant at about 70 mg/l for GAC residence times varying from 60 to 30 d and increased mildly afterwards. Since Reactor 1 treated acetic acid and phenol only, the

increase in the filtered effluent COD is attributed to an increase in the effluent concentrations of the biodegradable and poorly adsorbable compounds due to the reduction in the biological solids residence time (SRT) resulting from increasing GAC replacement rates. Obviously, the impact of GAC replacement rate on effluent quality is more drastic at SRTs close to the system critical SRT.

As depicted in Figs. 4(b) and (c), Reactors 2 and 3 followed the same trend of an initial increase in soluble effluent COD followed by a continuous drop until the end of the study. The initial increase in filtered effluent COD is due to the introduction of *o*-cresol into the feed on day. In these two reactors, as a result of the experimental protocol used, the effluent concentrations of *o*-cresol continuously decreased while the effluent concentrations of acetate and phenol increased with the reduction of the GAC mean residence time. It is therefore postulated that the increase in COD due to acetate and phenol was offset by the decrease in COD concentration due to *o*-cresol. It is worth mentioning that at low GAC residence times, the effluent COD concentrations due to the specific organic compounds represented a higher fraction of the filtered effluent COD than at high GAC residence times. Consequently, the COD due to the intermediate products of biodegradation may have contributed significantly to the effluent COD concentration at high GAC residence times.

Although soluble effluent COD from Reactor 2 was almost identical to Reactor 1 at low GAC residence times (16 d or less) corresponding to low effluent *o*-cresol concentrations, significant discrepancies were observed at high GAC residence times and cresol concentrations. Those differences, however, were mostly accounted for by the effluent cresol concentration. On the other hand, soluble effluent COD from Reactor 2 was generally underlying Reactor 3 COD at GAC residence times of 16 d and above, with only a minor fraction of the difference in COD contributed by the measured specific organics. It is therefore postulated that due to the more intense biological activity in Reactor 3 vis-a-vis Reactor 2, biodegradation intermediates contributed appreciably to the widely varying soluble effluent CODs measured from the two reactors.

Figs. 5(a)–(c) show the temporal variation in methane production rates measured in Reactors 1, 2 and 3 respectively. The potential methane, indicated by a broken line, represents the maximum methane that can be produced if the reactor converted all the influent acetate and phenol to methane gas. As apparent from the three figures, the three reactors converted more than 90% of the biodegradable influent COD to methane gas until a GAC replacement rate of 75 g/d was employed. After this replacement rate was used, the conversion efficiency of the influent COD to methane gas observed in Reactor 3 remained relatively constant at about 80% while it continued to decrease in Reactors 1 and 2 until it reached about 62% at the end of the study. The decrease in methane production rate with increasing GAC replacement rates is due to the increase in the effluent concentrations of the biodegradable constituents of the waste, namely acetate and phenol and the increasing removal of phenol by adsorption on activated carbon. Although the removal rate of phenol by adsorption on GAC at the 8 d GAC mean residence time was the same in Reactors 2 and 3, it represented a smaller fraction of the total biodegradable influent COD to Reactor 3 because of the higher organic loading rate applied to it. Despite the

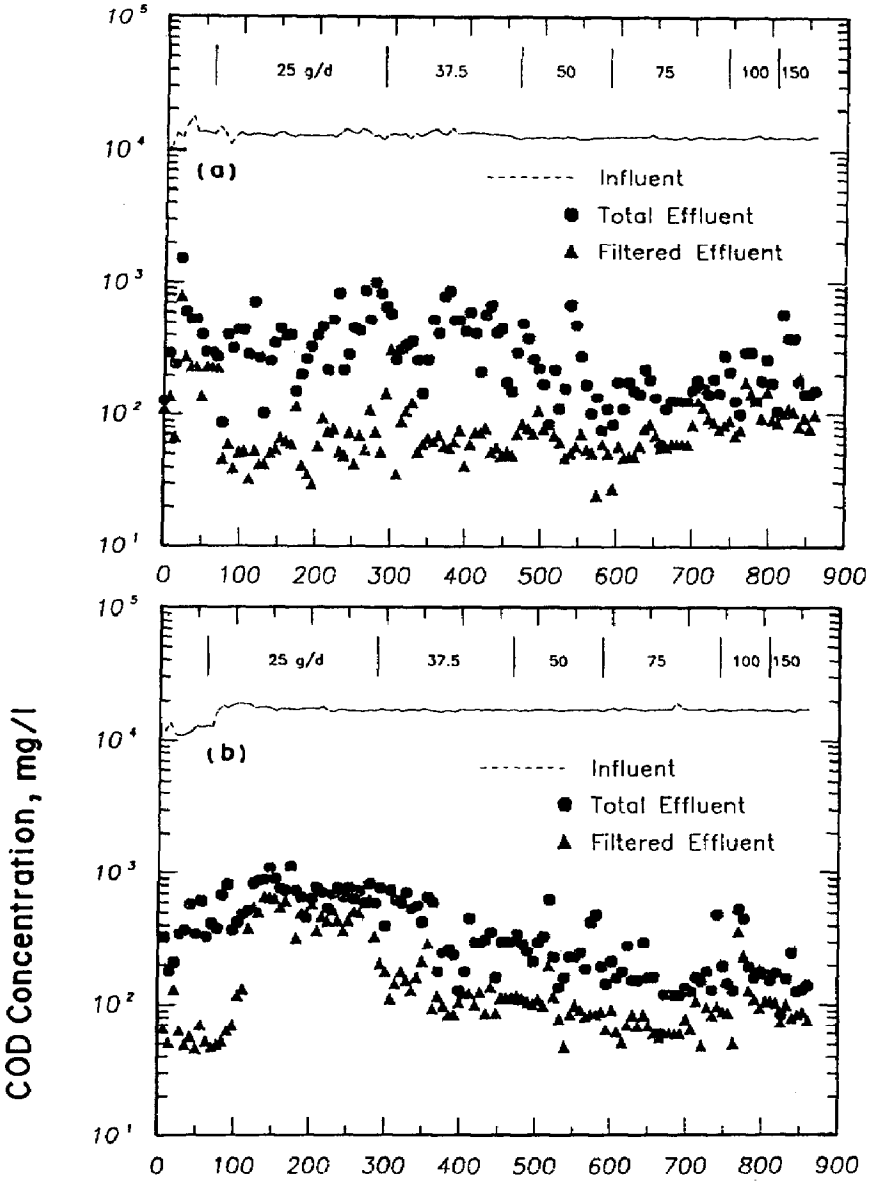

Fig. 4. Effluent COD variations in Reactor 1 (a), Reactor 2 (b), and Reactor 3 (c).

observed reduction in methanogenic activity in Reactors 1 and 2, the ability of the system to maintain significant anaerobic activity at GAC mean residence times as low as 8 d is remarkable.

On comparing the performance of Reactors 1 and 2, which were operated at identical biodegradable organic loading rates, with respect to effluent concentrations of selected organics and methane productivity, no inhibition of phenol degradation

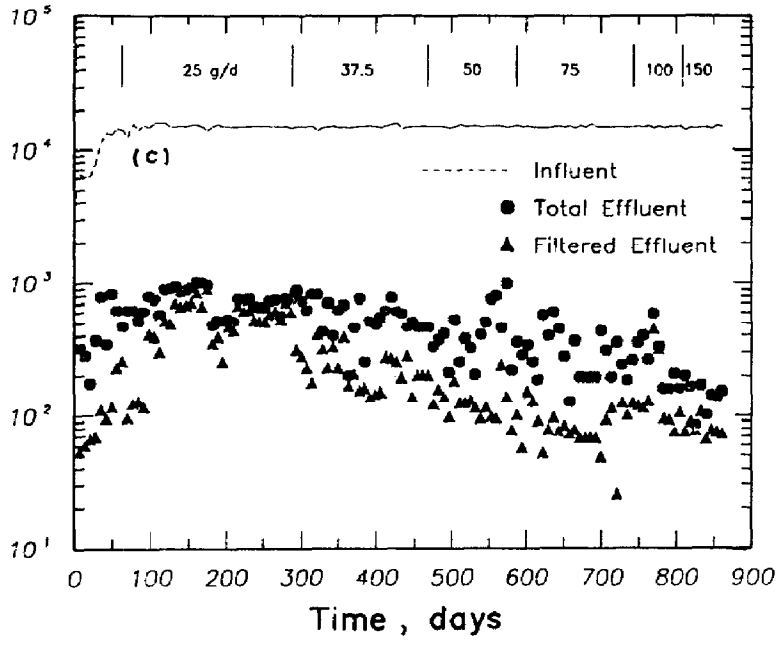

Fig. 4. Continued.

and methanogenesis, due to *o*-cresol was observed in Reactor 2 at any time during this study. On the other hand, the effluent concentrations of the biodegradable waste constituents i.e. acetate and phenol, from Reactor 2 were consistently lower, with the exception of the 100 g/d replacement period, than Reactor 3. At the 100 g/d replacement period, high biomass shear loss coefficients in Reactor 2, resulting in a significant reduction of the biological SRT below the GAC mean residence time caused a sharp increase in the effluent phenol and acetate concentrations from Reactor 2. Despite the relatively higher COD conversion efficiencies to methane gas in Reactor 3 shown in Fig. 5, some differences in the effluent COD from Reactors 2 and 3 were observed in this study. The excellent performance of Reactor 3 at a combined COD loading rate of 35.7 kg/m^3-d and an HRT of 0.5 d emphasizes the viability of the anaerobic GAC reactor for the treatment of high strength hazardous organic wastes.

The effluent concentrations of total suspended solids (TSS) and suspended carbohydrates from Rectors 1, 2 and 3 over the entire study are presented in Figs. 6(a), (b) and (c) respectively. The effluent TSS from the three reactors mostly ranged between 100–400 mg/l while the effluent carbohydrate fluctuated around the 20 mg/l level. As apparent from the figures, both parameters exhibited more scatter than the effluent concentrations of the specific organic compounds. Despite this scatter, some trends can still be observed. For example in Reactor 1, both the TSS and the suspended carbohydrate concentrations decreased continuously after employing the GAC replacement rate of 50 g/d to reach as low as 40 and 4 mg/l, respectively, at the end of the study. This decrease contributed to the drop in total effluent COD concentration discussed earlier. As apparent from Figs. 6(b) and (c), Reactors 2 and 3 exhibited the

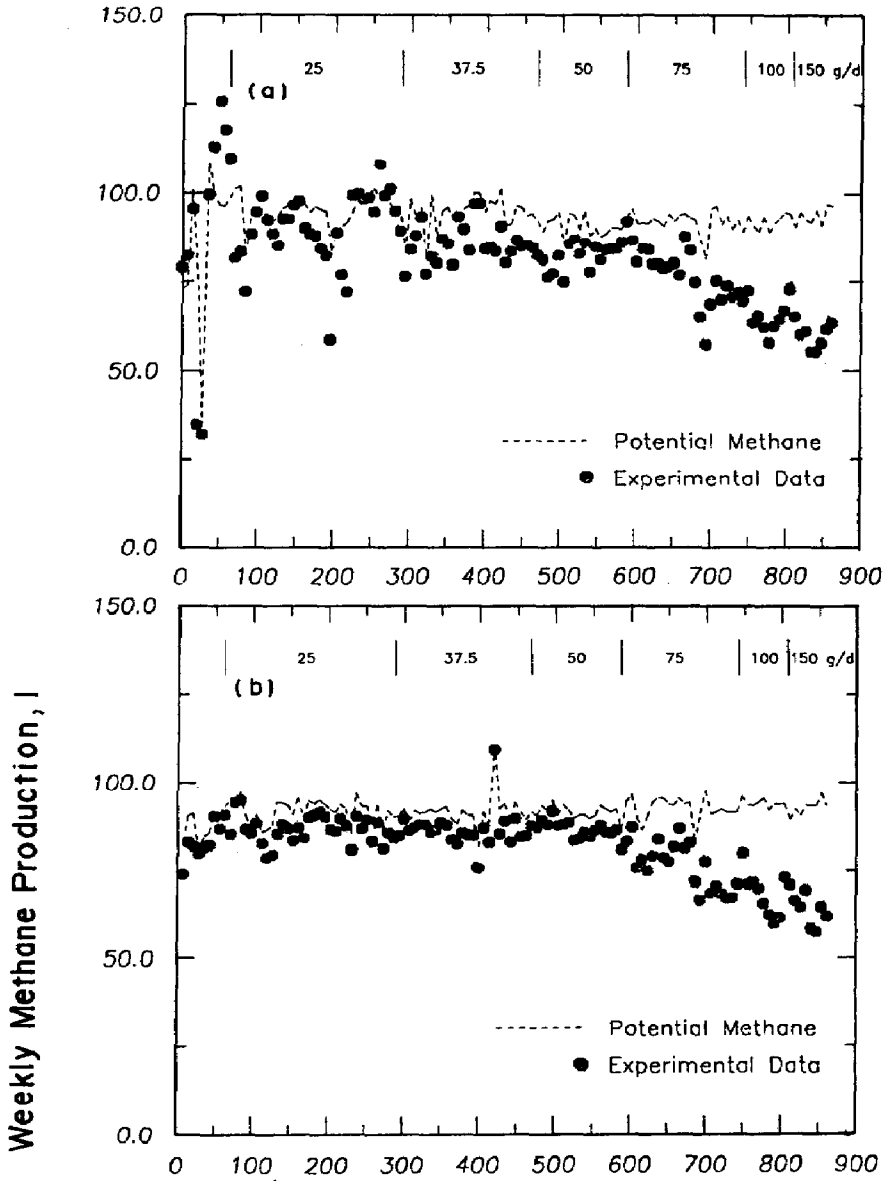

Fig. 5. Weekly methane production rates in Reactor 1 (a), Reactor 2 (b), and Reactor 3 (c).

same trend as Reactor 1. However, these trends cannot be explained confidently because of the limited knowledge of biomass shearing mechanisms. It is postulated that the increased availability of fresh attachment surface area at high GAC replacement rates may have caused the aforementioned reduction in effluent TSS.

The findings of this work not only point out to the interaction of adsorption and biodegradation in anaerobic GAC reactors but also underscore the importance of toxicity control by regular replacement of reactor GAC. The anaerobic reactor was

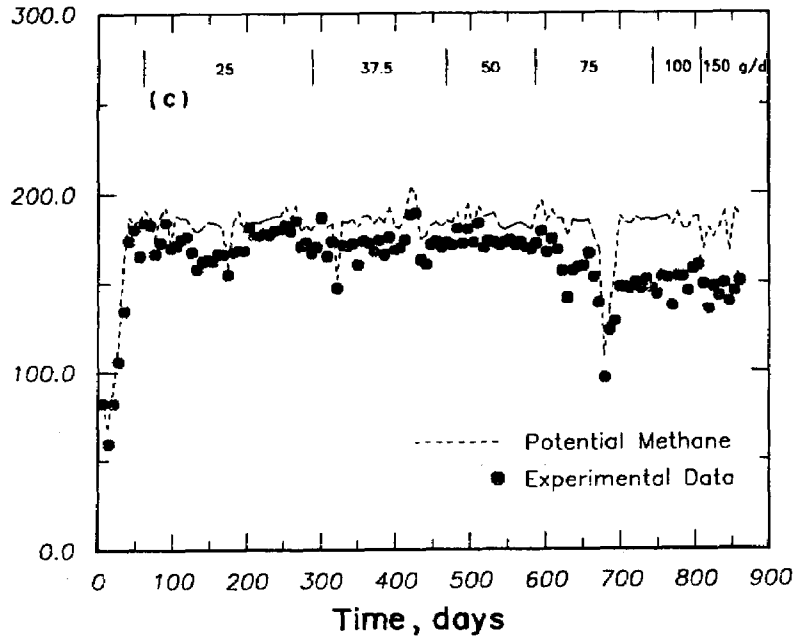

Fig. 5. Continued.

demonstrated to effectively treat a mixture of biodegradable and toxic compounds at a COD volumetric loading of 35.7 kg/m^3-d, a hydraulic retention time of 12 h and a GAC mean residence time of 8 d. Since it was shown that GAC replacement rates at which effluent concentrations of biodegradable adsorbable and nonadsorbable organics are minimum do not coincide with those construed optimal for removal of toxins, the results of this study incite practising engineers to carefully weigh the trade off between removals of biodegradable and nonbiodegradable waste constituents. It must be asserted that operation of an anaerobic GAC reactor to optimize removal of target organic compounds does not necessarily insure maximum reduction of total organics.

4. Summary and conclusion

Based on the findings of the research, the following conclusions can be drawn:

– The anaerobic GAC reactors affected more than 97% removal of the specific organic compounds with effluent acetate, phenol, and *o*-cresol concentrations ranging from 15 to 125 mg/l, 0.1 to 12 mg/l, and 0.6 to 50 mg/l respectively and over 99% removal of the influent COD. At high GAC mean residence times adsorbable biodegradable pollutants removal occurs by biodegradation, whereas at a low GAC mean residence times, adsorption becomes the dominant removal mechanism.

– COD to methane conversion efficiency dropped from over 90% in the reactors operated at a HRT of 1.0 d at the 60 d GAC mean residence time to about 62% at

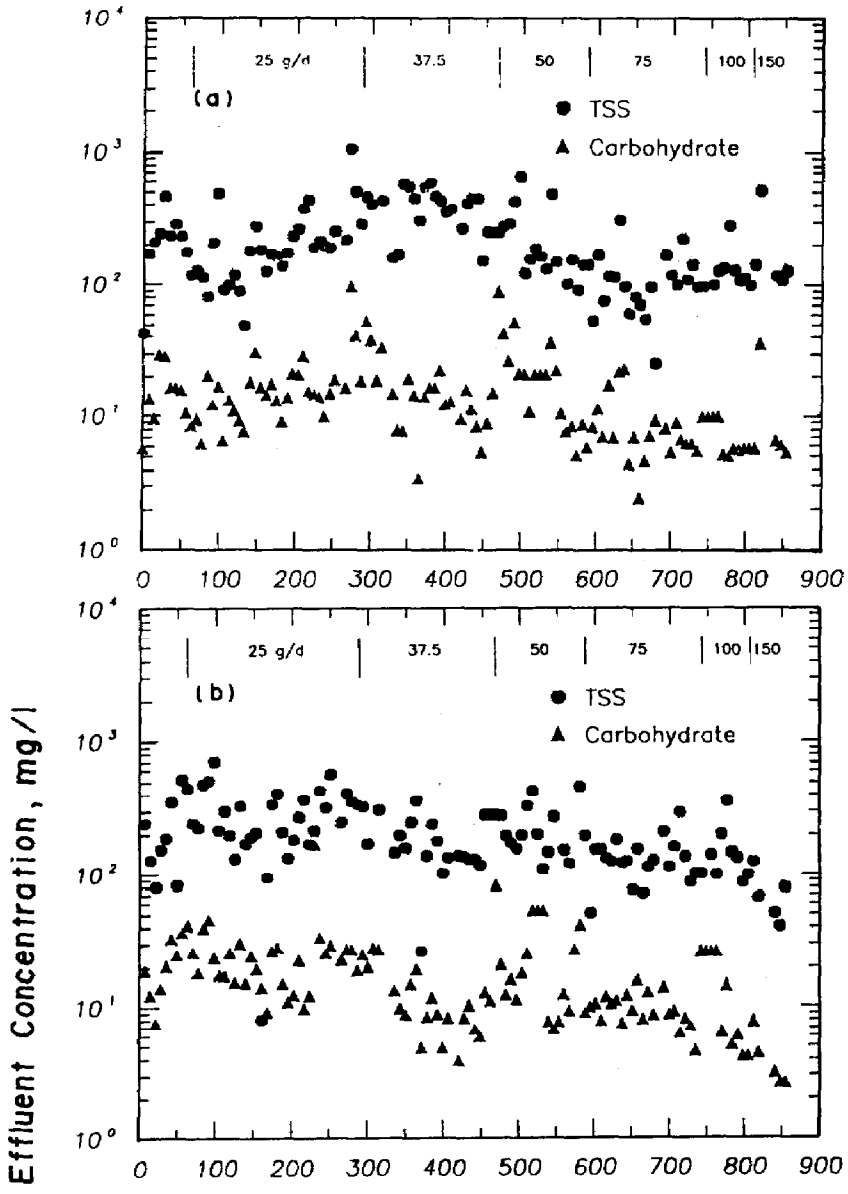

Fig. 6. Effluent TSS and suspended carbohydrate variations in Reactor 1 (a), Reactor 2 (b), and Reactor 3 (c).

a GAC mean residence of 8 d. The corresponding figures for the reactor operated at a HRT of 0.5 d are 90 and 80% respectively.

– The anaerobic GAC reactors can achieve successful treatment of toxic wastewaters at GAC mean residence times as low as 8 d and COD loading rates of 35.7 kg/m^3-d.

– No inhibition of phenol degradation and methanogenesis due to *o*-cresol loadings of 5.5 kg COD/m^3-d, was observed in this study.

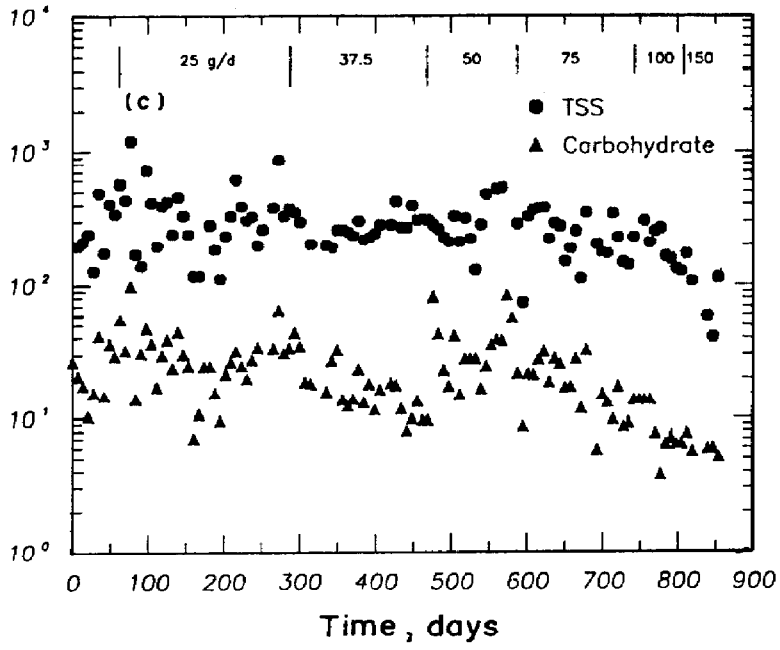

Fig. 6. Continued.

– Hydraulic retention time and biodegradable organic loading rate appeared to exert an appreciable impact on process performance. In fact, in terms of effluent quality, the reactor operated at an unexpanded empty bed HRT of 0.5 d was generally inferior to the other two reactors operated at a HRT of 1.0 d.

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