## ECONOMICAL AND ECOLOGICAL REMOVAL AND/OR RECUPERATION OF ORGANIC MATTER AND AMMONIUM FROM LANDFILL LEACHATE: A FULL SCALE CASE STUDY OF FLEMISH LANDFILLS.

Violet Oloibiri <sup>a,c,d</sup>, Michael Chys <sup>a,b</sup>, Wim Audenaert <sup>a,b</sup>, Kristof Demeestere<sup>c</sup>, Stijn WH Van Hulle <sup>a,b 1</sup>

<sup>a</sup>LIWET, Department of Industrial Biological Sciences, Ghent University, Campus Kortrijk, Graaf Karel de Goedelaan 5, B-8500 Kortrijk, Belgium.

<sup>b</sup>BIOMATH, Department of Mathematical Modeling, Statistics and Bioinformatics, Ghent University, Gent, Belgium.

<sup>c</sup>EnVOC, Department of Sustainable Organic Chemistry and Technology, Faculty of Bioscience Engineering, Ghent University, Coupure Links 653, B-9000 Ghent, Belgium.

<sup>d</sup>Kenya Industrial Research and Development Institute (KIRDI), P. O. Box 30650-00100, Nairobi, Kenya.

<sup>&</sup>lt;sup>1</sup>Correspondence to: SWH. Van Hulle, LIWET, Departement of Industrial Biological Sciences, Ghent University, Campus Kortrijk, Graaf Karel de Goedelaan 5, B-8500 Kortrijk, Belgium. E-mail: <u>stijn.vanhulle@ugent.be</u>, Tel: +32 (0)56 24 12 51

## Abstract

A major concern for landfilling facilities is the treatment of their leachate. To optimize organic matter and nitrogen removal from this leachate, the combination of several techniques is preferred in order to meet stringent effluent standards. For nitrogen removal, generally the nitrification-denitrification (N-DN) process is applied. However, this results in high costs as large amounts of oxygen and methanol are required to treat this nitrogen rich stream (1-5 gN/l) which contains little to no biodegradable organic matter. Two alternatives were investigated: autotrophic nitrogen removal (ANR) based on anammox and ion exchange. ANR with an N-DN polishing step resulted in similar removal performance, but reduced the operational cost from 0,57  $m^3$  to 0,36  $m^3$ . Using ion exchange a recuperation of over 60% of the ammonium could be achieved.

For organic matter removal, generally activated carbon adsorption is used. However, this also results in high costs as a large amount of activated carbon is necessary to remove the recalcitrant COD in the leachate (0,5-5 g/l). Pre-treating the leachate with coagulation flocculation resulted in a 10-fold increase of activated carbon operational time and a decrease of operational costs from 1,32 euro/m<sup>3</sup> to 0,88  $m^3$ , mainly because the COD load to the activated carbon columns was reduced and as such the life time of the columns was extended. Similar tests with ozone resulted in a decrease of operational costs to 1,2  $m^3$  and the conversion of 10% of the recalcitrant COD to BOD. This BOD can be used as carbon source for the preceding biological nitrogen removal. For both physical chemical treatment tests, the adsorption properties of the leachate did not change, nor was a deterioration of the final effluent quality after adsorption noticed.

Key words: landfill leachate treatment, organic matter, ammonium, cost evaluation

## Introduction

Landfilling remains the primary disposal method for municipal solid waste in developed and developing countries [1]. As a result of ground water intrusion, rainfall percolation and moisture present in the waste, deposits of toxic waste waters called landfill leachate are generated. Release of this leachate into the environment without proper treatment poses considerable risks to human and ecosystem health. The European Union council directive of 1999/31/EC and the Flemish environmental regulations VLAREM II require landfill operators in Belgium to undertake proper leachate treatment during the entire life cycle of a landfill to prevent any possible negative effects to the environment [2].

Several conventional as well as advanced treatment processes have been used to treat leachate. To meet the strict quality standards for the direct discharge of leachate into surface water, it is widely accepted that a combination of chemical (coagulation-flocculation, advanced oxidation processes (AOPs), physical (adsorption, membrane filtration, air striping) and biological steps are used [3]. The potential techniques for treatment of landfill leachate need to be evaluated based on their ability to reduce the pollutant load, available operational experience, energy requirements, process reliability and related environmental impacts [4]. To illustrate; the leachate treatment train consisting of air stripping, fenton oxidation, sequential batch reactor (SBR) treatment of leachate and sewage and final polishing using coagulation-flocculation managed to reduce the COD and NH<sub>3</sub>-N by 93 and 98% respectively [5]. The scheme also significantly improved the biodegradability of leachate (0.18 – 0.45) hence creating an opportunity for a recycle stream to the SBR unit. The competitiveness of this treatment train in terms of pollutant reduction as a criterion for evaluating and comparing treatment schemes is seen in different review papers [3,6,7].

Regardless of the aforementioned criteria, selection of the best available technique is based on their cost effectiveness. In view of their economy, several treatment plants incorporate a biological step [3,8]. Indeed, a survey of 166 leachate treatment plants by Alvarez-Vasquez et al [9] showed that 72% of the schemes had a biological method. Up to 60% of the reported biological methods were aerobic lagooning, activated sludge, and up-flow anaerobic sludge blanket. However, treatment of stabilized landfill leachate by the aforementioned methods is hampered by presence of bio-recalcitrant organics, high nitrogen concentrations and poor  $BOD_5/COD$  ratios (<0.2) [3]. Besides, additional carbon sources are required to aid the nitrification-denitrification process [10]. Full autotrophic nitrogen removal (ANR) processes are alternative biological methods for dealing with stabilized landfill leachate [11]. Depending on the operating conditions, up to 61% COD and 90% nitrogen removal can be achieved by ANR processes. Compared to nitrification-denitrification methods, ANR is known to consume 60% less oxygen and 40% less or no organic carbon [4] therefore, less operational costs.

AOPs are reported as the most effective methods in degradation of recalcitrant organic matter in stabilized landfill leachate [12,13]. However, their energy requirements are very high. To meet this demand, natural solar energy has been investigated as a cheaper alternative [14,15]. Yet the required compound parabolic collectors constitute a cost at least 24% of the total operating costs. In the combined treatment of landfill leachate using SBR, coagulation - floculation, Fenton and up flow anaerobic biological filters, 30% of the total treatment costs were attributed to reagents for the Fenton step [16]. A study [17] recommended the use of a cheap soil column as a final polishing step for pretreated leachate as opposed to activated carbon column which is more effective in COD,  $BOD_5$  and suspended solids removal. Leachate treatment costs are also affected by other conditions such as seasonal variations. For instance, in the dry season, the organic matter concentration in leachate increases [18,19]. This increases the chemical demand in case of chemical treatment which increases the operational costs as a result. The factors which affect leachate treatment can be summarized into a simple framework Figure 1 [20]. It shows that, at the macro level, treatment of landfill leachate is not only an environmental concern but also an economic one [16]. At the micro level, the environmental concerns are driven by the available technology and its efficiency, the operating conditions and environmental discharge standards. These three factors directly affect the quality of landfill leachate and impose a cost to the treatment of landfill leachate [16,21].



Figure 1: The factors directly affecting landfill leachate quality and their relationship with environmental concerns and costs

In Flanders (the Northern part of Belgium), landfill operators have minimum or no control over strict discharge standards and operating conditions. Therefore, to alleviate the environmental concerns, associated with landfill leachate, landfill operators must constantly review their treatment processes. In certain cases, incorporate other techniques or totally overhaul their system. Notwithstanding, the choice must allow the operators to comply to the environmental discharge standards at the lowest possible costs. From this perspective a case study was done to investigate how the choice of technology affects the leachate quality and treatment costs. Using the leachate treatment scheme at intergemeentelijke maatschappij voor openbare gezondheid (IMOG) [10,11] - a landfill facility in Flanders - as a model, the effect of replacing classic nitrification-denitrification (N-dN) with autotrophic nitrogen removal (ANR). Additionally the incorporation of different techniques namely ozonation, fenton and coagulation flocculation in the treatment chain, allows to meet strict standards but also affects the treatment costs. As an alternative to IMOG's existing treatment train, the potential of ANR, ozonation and activated carbon adsorption to treat stabilized landfill leachate and their resulting costs will be presented.

## Methodology

## IMOG Moen landfill site and leachate characteristics

The leachate used in the experiments was sampled from IMOG. The sampling and storage procedures are outlined in Chys et al. [10]. IMOG is a landfilling facility located in Moen (Belgium). The facility houses four landfills and covers approximately 27 acres. The facility generates  $150 \text{ m}^3/\text{day}$  of landfill leachate. The leachate first goes through filtration to remove soil particles and oil followed by biological treatment (nitrification-denitrification with methanol addition) using SBR and reed beds to remove ammonium nitrogen. Final polishing using granular activated carbon (GAC) is done to remove bio-recalcitrant compounds before final discharge. Characteristics of the raw and biological leachate used in the experiments are summarized in Table 1.

Parameter	Raw Leachate	SBR treated leachate
pH	8.13	8.2 - 8.5
COD(mg/L)	1185	706 - 2260
$BOD_5(mg/L)$	189	50
BOD <sub>5</sub> /COD	0.15	0.03
$NH_4^+$ - N(mg/L)	244 - 627	2.42 - 9.22
$NO_3 - N(mg/L)$	7.7	3.9 – 19.5
$NO_2^ N(mg/L)$	10	0.33 - 0.66
$UVA_{254}$ (cm <sup>-1</sup> )	NA	5.98 - 8.53
Conductivity (µS/cm)	10002	6870
NIA Net Assoilable		

Table 1. Characteristics of the raw and biological leachate used in the experiments in this study

NA: Not Available

## **Experimental design**

Data used in this study was selected from seven studies carried out previously by our research group. To compare the cost implications and efficiency of different biological techniques in treating stabilized landfill leachate, prefiltered landfill leachate was treated using ANR instead of N-dN. The ANR process was accomplished via partial nitritation and ammonium oxidation (anammox) in a single reactor. Descriptions for the reactor set-up and operating conditions are outlined in [11]. Incorporation of other techniques into IMOG's existing treatment train to meet the discharge standards was evaluated. The biological treated leachate was further treated separately using coagulation-flocculation, Fenton oxidation and ozonation, after which the effluents underwent activated carbon adsorption. Amongst several chemicals, ferric chloride (FeCl<sub>3</sub>) was found to be a good coagulant for the coagulation process. The optimization experiments are reported by [22]. The pH variations and optimum concentration for Fe<sup>2+</sup> and H<sub>2</sub>O<sub>2</sub> addition in the Fenton experiments are described in [10]. Descriptions for the ozonation process and test results with leachate of different concentrations are found in [23] and [10]. As an alternative treatment sequence, the prefiltered leachate was treated using ANR followed by ozonation and adsorption with granular activated carbon. This was done under the conditions outlined in [10,24]. To reduce costs associated with addition of an external carbon source, ozonated leachate was partially recirculated to the ANR reactor [24].

Evaluation of the biological techniques was based on COD and nitrogen removal. The efficiency of ozonation, and Fenton to reduce leachate quality to the required discharge limits was based on COD and increase in biodegradability (BOD<sub>5</sub>/COD). COD reduction and sludge production were used to study coagulation flocculation.

#### **Cost evaluation**

The economic feasibility of each technique was based on the reagents used. This is because several researchers noted that reagents costs contribute 50 - 89% to the total operating costs [14,16,21,25]. For biology and ozonation, energy costs were found to be significant [13] hence were taken into account. The energy costs were based on the electricity consumption. Only the disposal costs for sludge produced in the biological step were taken into consideration. The disposal costs of sludge generated during coagulation flocculation and Fenton were not taken into account as this can potentially be recycled to the landfill. In a detailed study, this disposal costs will have to be taken into account.

#### **Results and Discussion**

#### Changing one biological mechanism for another: nitrification-denitrification vs ANR

Nitrification-denitrification (N-dN) is a robust biological technique that removes nitrogen from landfill leachate. From the leachate characteristics shown in Table 1, N-dN was efficient in reducing the ammonium concentration in landfill leachate below the Flemish discharge limits (5 mg/L). Values above 5 mg/L are from samples collected during the winter period when biological reaction rates are lowered by cold temperatures. The poor COD removal or lack thereof is an indication that the denitrification stage was driven by an external source of carbon. IMOG, like most leachate treatment plants employs methanol as an external carbon source. Therefore, assuming a stoichiometric methanol dosage of 2.47g CH<sub>3</sub>OH/ g N<sub>NO3</sub><sup>-</sup> (3.7g COD/g N<sub>NO3</sub><sup>-</sup>) to achieve 100% total nitrogen removal from a leachate stream containing 60 kg N/d (influent concentration 0.4 kg N/m<sup>3</sup> \* flow rate 150 m<sup>3</sup>/d), the methanol costs were 0.404  $\notin$ m<sup>3</sup>. The costs associated with electricity consumption during aeration and sludge disposal were 0.088  $\notin m^3$  each. As such, the total operating costs of the nitrification denitrification process with N-dN is  $0.58 \notin m^3$  (31,755  $\notin$ year). As predicted, up to 71% of the total operation costs was spent on methanol which is the main reagent. Similar observations were reported by Cassano et al [21]. Methanol consumption in the treatment of landfill leachate by sequencing batch biofilter granular reactor accounted for 60% of the total operating costs. In comparison only 18% and 7% of the total costs were attributed to aeration and sludge disposal respectively. To achieve the discharge limits for COD, further treatment of the biological effluent using GAC adsorption cost IMOG a further 1.32 €m<sup>3</sup> (72,270 €year). Therefore, the total treatment costs for leachate using N-dN and GAC (this combination is further denoted as A0) is 1.9 €m<sup>3</sup> (104,025 €year)

To reduce costs, it is important that the external carbon source consumed is reduced while increasing the nitrogen removal efficiency. To achieve this, ANR was used which involves partial oxidation of ammonium into nitrite to achieve a theoretical nitrite ammonium ratio of 1:1 [26] and the anammox process where ammonium is used as an electron donor and nitrite as an electron acceptor to produce nitrogen gas [4,27]. Lab scale operations of the ANR reactor is sensitive to changes in hydraulic loading rate (HRT), nitrogen loading rate, dissolved oxygen and temperature. At HRT of 2 days, the nitrogen removal efficiency varied between 45% and 14%. Instances of poor performance corresponded with reduced nitrogen loading rates (153 mg N/L.d) and nitrite concentrations of 253 mg/L in the effluent. Nitrite concentrations above 100 mg/L are known to inhibit the anammox process [28]. Increasing the HRT to 3 days raised the nitrogen removal efficiency to 72%. A decrease in nitrite concentrations in the effluent below 20 mgN/ L was also observed. Overall, optimizing the HRT (3 days), nitrogen loading rate and dissolved oxygen concentrations ( $0.3 - 0.5 \text{ mg O}_2/L$  in the reactor led to 55% total nitrogen removal (Gao et al., 2014). Costs estimations for the removal of nitrogen by ANR only take into account oxygen consumption costs as methanol is not needed in the process and sludge production is negligible. Considering the stoichiometric balance between ammonium and oxygen in ANR, leachate flow rate of 150  $\text{m}^3/\text{d}$ influent ammonium concentration of 400 mg/L and nitrogen loading rate of 60 kg N/d, the aeration costs were 0.04 €m<sup>3</sup>. This results in total operating cost of 2190 €year. Aeration costs of the ANR process are much less than the costs incurred in the N-dN process. Furthermore, up to 30,000 €year can be saved by using the ANR for nitrogen removal instead of N-dN. If only a limited amount of nitrogen is removed by ANR (for example 40%) than part of the removal should be attributed to N-dN (in this case 60%) [11]. Therefore the additional costs for methanol addition and aeration are  $0.6 * 0.58 \notin m^3 = 0.348 \notin m^3$ . Total operational costs for combined ANR and N-dN is  $(0.4*0.04) + 0.348 = 0.36 \notin m^3$  (19,929  $\notin$  year). From the yearly costs, combined ANR and denitrification is 38% cheaper than the full N-dN process. Additionally COD removal can be achieved. If the GAC treatment costs for the two biological techniques are the same (1.32  $m^3$ ), then the total costs for leachate treatment using ANR and GAC (further denoted as A1) is 1.36 €m<sup>3</sup> (74,460 €year)

# Incorporation of techniques into the existing treatment process (between biological treatment and adsorption)

Post treatment of biologically treated landfill leachate before final polishing with GAC is a viable option for achieving the set limits for discharge of treated landfill leachate and improvement of adsorption capacities of activated carbon.

Using data presented in [29], ozonation of landfill leachate resulted in increased COD removal as the ozone dosages increased. At a maximum dosage of 4.84 g  $O_3/g$  CODo, 44% COD removal was achieved. Using the energy (1,44  $\notin$ kgO<sub>3</sub>) and oxygen prices (0,84  $\notin$ kg O<sub>3</sub>) outlined by [10] the total costs for treating leachate (112 mg/L COD) at this dose was 0.78  $\notin$ m<sup>3</sup> (energy) + 0.455  $\notin$ m<sup>3</sup> (oxygen) = 1.23  $\notin$ m<sup>3</sup> (66,7668  $\notin$ year). On the contrary, treating undiluted leachate (COD<sub>0</sub> 724 mg/L) at a more economically feasible ozone dose of 0.14 g O<sub>3</sub>/g COD<sub>0</sub> [10] resulted in up to 10% COD removal with 0.14  $\notin$ m<sup>3</sup> and 0.08  $\notin$ m<sup>3</sup> spent on energy and oxygen for ozone generation respectively. GAC adsorption of ozonated effluent resulted in improved adsorption capacities and breakthrough time of the activated carbon column [10,22]. Consequently, GAC treatment of ozonated leachate costs for treatment of leachate using N – dN, ozonation and GAC adsorption (further denoted as A2) is 0.58 +0.22+0.95 = 1.76  $\notin$ m<sup>3</sup> (96,507  $\notin$ year). Addition of ozonation to the IMOG leachate treatment train reduces the operation costs by 7%. The UV absorbance of leachate at longer (500 nm) and shorter (254 nm and 350 nm) wavelengths also decreased during ozonation.

Fenton oxidation is another AOP which was investigated as a potential alternative to ozonation. It was chosen because it is capable of removing a wide variety of compounds from landfill leachate [30]. Experiments by [10] show that dosage of Fe<sup>2+</sup> and H<sub>2</sub>O<sub>2</sub> at 1117 mg/L (1.4 g Fe<sup>2+</sup>/g COD<sub>o</sub>), 1020 mg /L (1.3 H<sub>2</sub>O<sub>2</sub>/g COD<sub>o</sub>) at pH 6 give the best removal of organic matter from landfill leachate. At these conditions, up to 67% UV absorbance at 254 nm and 63% COD reduction was achieved. Taking into account the chemical doses and cost of FeSO<sub>4</sub>.7H<sub>2</sub>O which was the Fe<sup>2+</sup> source (0.33  $\notin$ kg) and H<sub>2</sub>O<sub>2</sub> (0.385  $\notin$ kg) the operating costs incurred include 0.37  $\notin$ m<sup>3</sup> for FeSO<sub>4</sub>.7H<sub>2</sub>O and 0.79  $\notin$  m<sup>3</sup> for H<sub>2</sub>O<sub>2</sub>. At least 0.13  $\notin$  m<sup>3</sup> was also used in lowering the pH with HCl. With these costs, Fenton oxidation was found to be much more expensive than ozonation. However, calculating costs with regards to the COD removed, treatment of landfill leachate using Fenton oxidation cost 2.5 € g COD removed compared to ozonation which used 3.1 €/g COD removed [10]. The Fenton oxidation effluent should be further treated using GAC. During the test [10] the effluent COD from the GAC column did not go above the environmental discharge limit for COD even after 14 bed volumes. For this reason, GAC adsorption of Fenton oxidation effluent cost was estimated to be low (0.09 €m<sup>3</sup>). In total, treatment of landfill leachate using N-dN, Fenton oxidation and GAC (further denoted as A3) is 1.96 €m<sup>3</sup>(107,310 €year). Incorporating Fenton oxidation into the IMOG treatment process proved to be 3% more expensive than the current process. Similar observation were reported by [25] where by treatment of landfill leachate by returned activated sludge only cost about 100,000 € while the addition of a Fenton oxidation step increased the operation costs to about 500,000 €

At pH 6, coagulation is the dominant mechanism of action in Fenton oxidation. Therefore, to investigate the role of coagulation only on COD removal and the respective costs, ferric chloride (FeCl<sub>3</sub>) and poly aluminium chloride (PACl) were used to treat landfill leachate. Generally, increase in coagulant concentration promoted higher reductions in leachate COD and UV absorbance [22]. In experiments without pH reduction, better COD (66%) and UV absorbance (88%) reductions were obtained with 1 g FeCl<sub>3</sub>/g COD<sub>o</sub> compared with 44% COD and 72% UV absorbance removal at 1 gPACl /gCOD<sub>o</sub> [31]. Additionally, sludge produced during FeCl<sub>3</sub> treatment had better settling properties as shown by the low sludge volume index (154 mL/g). The SVI of PACl was 250 mL/g. As such further coagulation experiments with pH reduction were conducted using FeCl<sub>3</sub>. At pH 6, influent COD concentration was reduced by 59% using 1 gFeCl<sub>3</sub>/g COD<sub>o</sub> [10]. This removal efficiency is comparable to 63% obtained during Fenton oxidation. Thus it can be assumed that the GAC treatment costs for coagulated effluent will be comparable to GAC treatment of Fenton oxidation effluent. Considering FeCl<sub>3</sub> cost of 0.625  $\notin$ kg a coagulation cost of 0.81  $\notin$ m<sup>3</sup> was obtained. Similarly to Fenton treatment, a low cost for GAC treatment (0.09  $\notin$ m<sup>3</sup>) was used. The total costs for nitrification-denitrification, coagulation and GAC adorption (further denoted as A4) of landfill leachate is 1.48  $\notin$ m<sup>3</sup> (81,030  $\notin$ year). Evaluation of the costs show that addition of coagulation in the IMOG process reduces the total treatment costs by 21%.

#### Alternative treatment configurations

Use of a completely different process to treat leachate from IMOG is another viable option to meet the effluent discharge standards. In the aforementioned studies, ANR and ozonation gave good nitrogen and COD removals in addition to lowering the leachate treatment costs. As such the applicability of ANR, ozonation and GAC (further denoted as A5) in reducing the pollutant load of IMOG landfill leachate was investigated.

Ozonation of ANR effluent at ozone concentration at dosages between 0.1 and 0.3 gO<sub>3</sub>/g CODo increased the biodegradability of leachate from 0.001 -0.002 to 0.01-0.06 [24]. This occurred as a result of COD reduction and subsequent increase in BOD<sub>5</sub>. The corresponding costs for ozonation of ANR effluent were in the range 0.19 - 0.69  $m^3$ . Recirculation of ozonated ANR influent to the ANR set up at a ratio of 1:9 (ozonated ANR leachate: raw leachate) improved the COD removal efficiency of ANR from approximately 5% to 12%. Subsequent ozonation of this combined leachate significantly lowered the ozonation costs to a minimum of 0.06  $m^3$ . Though higher recirculation ratios 1:3 and 1:1 provided better COD removal (28% and 40% respectively), the ozonation costs were higher.

Economic analysis of post treatment of ozonated effluent with GAC show that the treatment costs are significantly reduced to 0.88 -1.2  $m^3$  from 1.32  $m^3$  as a result of enhanced GAC adsorption properties. On average, leachate treatment with ANR, ozonation and GAC is 0.04+0.19+0.88 = 1.11  $m^3$  (60,772  $m^3$ ) (60,772  $m^3$ ) comparing the treatment costs between the existing process at IMOG and the proposed alternative process, we found that the alternative process was 41% cheaper and better removals of COD (78%) and nitrogen (82%) could be obtained

The different treatment options are summarized in Figure 2.



Figure 2: the proposed alternative processes for treatment of landfill leachate in comparison with the existing treatment process (for treating  $150 \text{ m}^3/\text{d}$ ).

## Conclusions

As illustrated in this evaluation, several strategies are available for landfill operators to meet the environmental discharge limits for treated leachate while keeping costs at a minimum. Proposed alternative process should take into account the already existing infrastructure to reduce capital costs for installation of other steps. From Figure 2 it can be concluded that changing the working mechanism of a single step in the treatment train e.g. in biology from N-dN to ANR can significantly reduce the costs involved in biological treatment of leachate. However, this

has no impact on the adsorption costs. Interestingly, post treatment of biological effluent before GAC adsorption does not necessarily reduce treatment costs as shown by the treatment process A3. Of the three techniques used in post treatment of biological effluent, ozonation was the cheapest though Fenton oxidation and coagulation drastically reduced the costs associated with GAC. Though ozonation in alternative process A3 might appear cheaper, the long term impact on activated carbon consumption should be monitored as this might render it costly. Therefore, to use alternative process A3, A4 and A5, landfill operators should consider long term operation costs and other costs for e.g. activated carbon regeneration and sludge disposal. Of all the process, alternative process A5 with ANR, ozonation and GAC was the cheapest (Figure 2). Given that a GAC column is already in use at IMOG and ANR can be done in the available SBR reactors with the only capital costs incurred are those for ozonation.

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