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Landfill leachate treatment by the combination of physicochemical methods with adsorption process

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ABSTRACT

The pretreatment with lime, ammonia stripping, chemical coagulation and activated carbon adsorption were used respectively for Bursa region leachate in this study. It was used that the first alternative, pretreatment with lime+ammonia stripping+neutralization were operated. This alternative achieved 19% COD removal and performed the capital and annual operating costs of \$175 m^{-3} and \$5 m^{-3}, respectively. In this study, the second alternative included chemical coagulation+ammonia stripping+granular activated carbon (GAC) adsorption. This alternative flow scheme achieved to produce an effluent COD of 160 mg L^{-1}, which complied with Turkish discharge standard for receiving media and capital&annual operating cost were estimated to be \$383 m^{-3} and \$18 m^{-3}, respectively. Consequently, the second alternative was determined to be appropriate according to discharge standard. But, it's capital and operating costs are very high for discharge to surface water. The following pretreatment with lime, co-treatment with municipial waste water is appropriate for lower treatment costs. Recommended this solution shows suitable results about treatment ammonia and heavy metals. Furthermore this solution can help that the developing countries adopt landfill leachate treatment.

Key Words: Adsorption, landfill leachate, physicochemical treatment

INTRODUCTION

Wang et al. (2002) indicated that the municipal landfill leachate has been one of the major problem for environment because of high organic, inorganic and heavy metal content and toxicity characteristics. In the operations, leachate treatment is both a very difficult and expensive process. Although, young leachate can be treated easily by biological treatment, COD removal efficiency are usually low due to high ammonium ion content and the presence of toxic compounds such as metal ions (Sletten et al., 1995; Amokrane et al., 1997; Irene and Lo, 1997; Chiang et al., 2001). The advanced treatment methods such as adsorption and UV/H_2O_2 can be necessity for treatment of landfill leachate because of toxic and recalcitrant constituents. Therefore, in this study the combination of physicochemical methods with granular activated carbon adsorption were expressed as a solution.

The any single method is no available for environmentally friendly and economically. There will be many other studies concerning the best available technology providing both maximum treatment efficiency and optimum cost.

The landfill leachate treatment methods are physical, chemical and biological ones which are used in combinations. Air-stripping , adsorption, membrane filtration are major physical leachate treatment methods (Amokrane et al., 1997; Bohdziewicz et al., 2001; Morawe et al., 1995; Trebouet et al., 2001), coagulation-flocculation, chemical precipitation, chemical and electrochemical oxidation methods are the common chemical methods used for the landfill leachate treatment (Amokrane et al., 1997; Ahn et al., 2002; Chiang et al., 2001; Lin and Chang, 2000; Steensen, 1997; Marttinen et al., 2002). In the landfill leachate treatment, a 66% COD and 50% ammonia removal were obtained by nanofiltration (Marttinen et al., 2002). Li et al. (1999) reported that ammonium removal by chemical precipitation can be achieved. Di Palma et al. (2002) used evaporation and reverse osmosis for the treatment of industrial landfill leachate. Trebouet et al. (2001) expressed that using nanofiltraton and chemical coagulation, COD content of leachate reduced less than discharge standards. Kargi and Pamukoglu (2003), Koh et al. (2004), Wang et al. (2002) and Rivas et al. (2003) also successfully applied combined processes; coagulation-flocculation + biological treatment; photochemical oxidation + activated sludge; Fe(III) chloride coagulation + photo-oxidation; and ozonation + adsorption. The several researchers in the literature investigated efficiency of ozonation alone for treating landfill leachate (Baig et al., 1999; Kuo, 1999; Silva et al., 2004; Steensen, 1997). Silva et al. (2004) achieved that COD and color at an ozone dose of 3 g L^{-1} was removed 48% and 87%, respectively.

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In the literature, combined chemical and biological treatment of landfill leachate has also been investigated. Geenens et al. (2001) improved efficiency of biological treatment using ozone pre-treatment before biological treatment of landfill leachate. They reported that pretreatment with ozonation decreased COD/BOD ratio from 16 to 6.

In this study focuses on the investigation of the efficiency and economy of treatment of landfill leachate from a municipal waste dump apart from all other similar studies. Alternative I and II mentioned bottom were compared about efficiency and economic of treatment.

- *Alternative I:* Pretreatment with lime + ammonia stripping + neutralization (Figure 1);
- *Alternative II:* Chemical coagulation + ammonia stripping + granular activated carbon (GAC) adsorption (Figure 2).

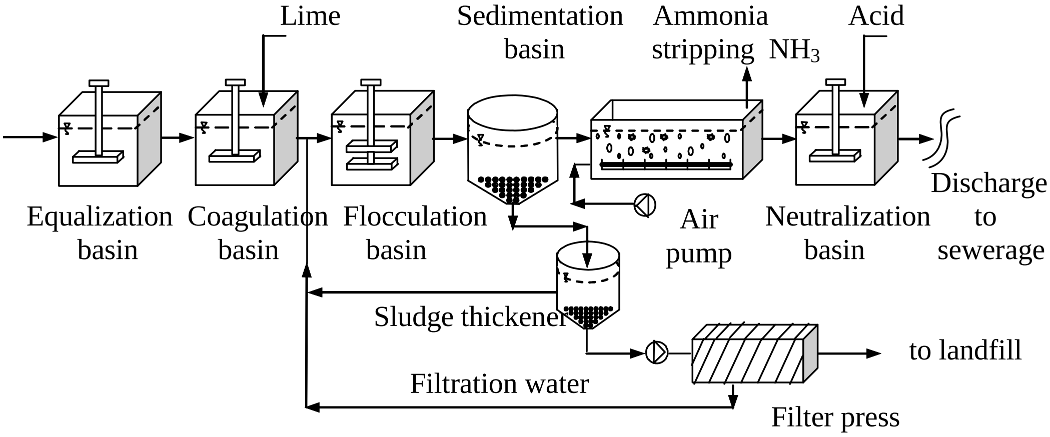

Figure 1. Schematic of lime pretreatment and ammonia stripping of leachate

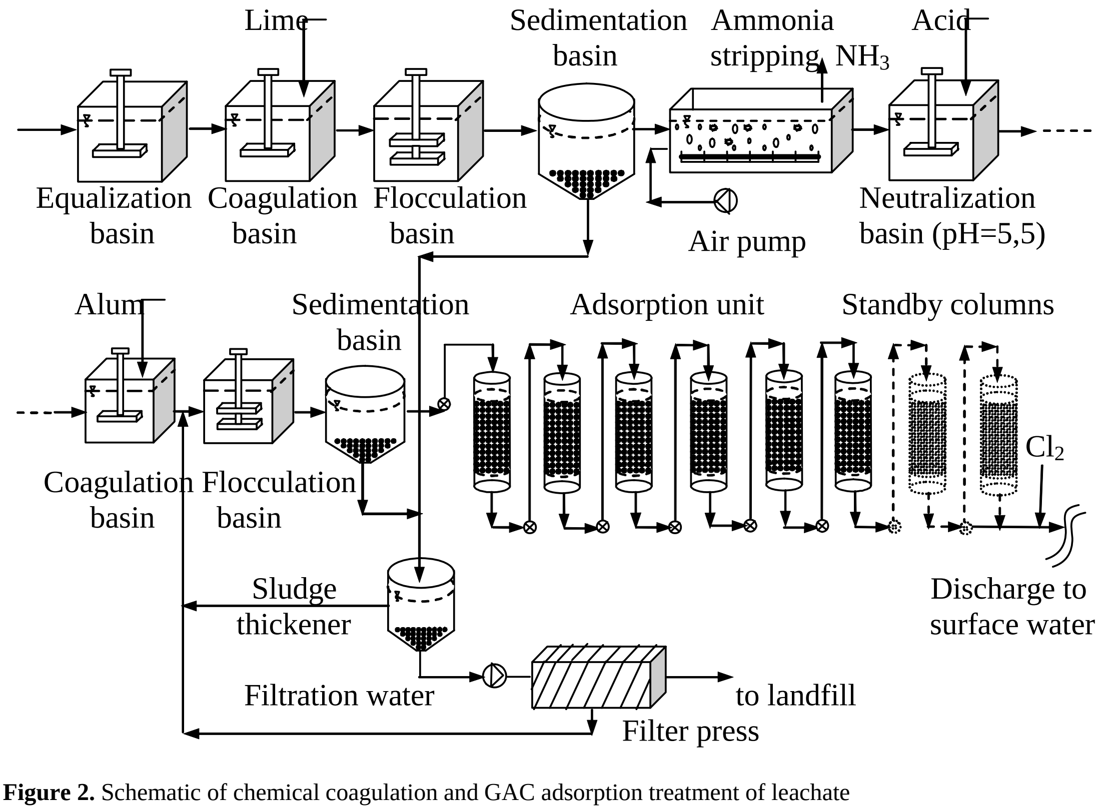

Figure 2. Schematic of chemical coagulation and GAC adsorption treatment of leachate

MATERIALS AND METHODS

Wastewater Sample

In this study, the leachate sample was provided from a landfill place in Bursa region where is in the west of Turkey. This landfill place with average leachate generation of 350 m^3/day has been served since approximately 10 years. From a wastewater pond in landfill place, obtained leachate sample was 50 liters. The leachate to result from landfill was stored at 4°C in this wastewater pond. In addition continuous characterization study was also carried out and results of years 1998 through 2000 are given in Table 1.

Table 1. Characteristics of leachate samples in 1998, 1999 and 2000 years

Parameter	Unit	Average of Year 1998	Average of Year 1999	Average of Year 2000
BOD_5	mg L^{-1}	22904	11894	15373
COD	mg L^{-1}	32957	19048	24373
SS	mg L^{-1}	1825	1429	2024
pH	-	7.1	7.1	7.4
Fe^{+2}	mg L^{-1}	100.3	42.5	32.6
Cu^{+2}	mg L^{-1}	15.2	11.6	15.5
Cd^{+2}	mg L^{-1}	2.82	0.06	0.16
Zn^{+2}	mg L^{-1}	49.8	49.6	27.2
Pb^{+2}	mg L^{-1}	56.7	7.03	9.18
Cr^{+6}	mg L^{-1}	7.0	6.6	7.4
F^-	mg L^{-1}	14.1	4.1	7.7
Phenol	mg L^{-1}	89	-	-
$SO_4^=$	mg L^{-1}	-	-	-
Sulfide	mg L^{-1}	-	-	-
NH_4^+-N	mg L^{-1}	72.8	-	-
Total Nitrogen	mg L^{-1}	-	-	-
Total Alkalinity	mg $CaCO_3$ L^{-1}	-	-	-
Total Chromium	mg L^{-1}	8.9	7.4	8.8
Total Cyanide	mg L^{-1}	8.10	4.36	3.0
Total Phosphorus	mg L^{-1}	38.4	-	-
Conductivity	mS	4765	-	349

- average of 68 samples collected in 1998
- average of 49 samples collected in 1999
- average of 368 samples collected in 2000

Analytical Procedures

The suspended solids (SS), pH, alkalinity and chemical oxygen demand (COD) etc. for leachate sample were determined consider Standard Methods (APHA, 1998) and characteristics of leachate sample are given in Table 2. Using 0.45 µ filter paper, supernatant was filtered and analysed by Atomic Absorption spectrometer (ATI Unicam 929 AA Spectrometer) for heavy metal analysis.

Pretreatment with lime and ammonia stripping

The leachate sample pH was increased to 11 by pretreatment with lime (10% $Ca(OH)_2$ by weight). After that sample was precipitated approximately 2 hours period and for COD, SS and heavy metals the supernatant was analyzed. The aeration (15 L air/min) was applicated in the supernatant throughout 24 hours after pretreatment and ammonium concentration was decreased from 1140 mgL^{-1} to 451 mg L^{-1} by this aeration. With acid, effluent was neutralized until pH demand of operation. Table 2 shows leachate sample characteristics after pretreatment.

Table 2. Characteristics of raw sample and treated sample with lime ($Ca(OH)_2$, 10% by weight)

Parameter	Unit	Raw Sample	Treated Sample	Removal (%)
BOD_5	mg L^{-1}	13450	-	-
COD	mg L^{-1}	23700	19200	19
SS	mg L^{-1}	1850	345	82
pH	-	7.57	1.84	-
Fe^{+2}	mg L^{-1}	23.2	2.65	89
Cu^{+2}	mg L^{-1}	10.4	2.5	76
Cd^{+2}	mg L^{-1}	0	0	-
Zn^{+2}	mg L^{-1}	59.4	21.2	64
Pb^{+2}	mg L^{-1}	3.1	0.5	84
Cr^{+6}	mg L^{-1}	7.6	5.95	22
F^-	mg L^{-1}	3.7	-	100
S^{-2}	mg L^{-1}	0.4	-	100
NH_4^+-N	mg L^{-1}	1140	451	60
Total N	mg L^{-1}	4540	4230	7
Total Chromium	mg L^{-1}	9.8	6.2	37
Total Cyanide	mg L^{-1}	1.4	0.75	46
Total Phosphorus	mg L^{-1}	20.8	3.4	84

Chemical Coagulation

Using Jar Test equipment in the physicochemical treatment experiments were investigated the effect of $Al_2(SO_4)_3.18H_2O$ (supplied from Merck) at various dosages on removing the COD and turbidity content of leachate. The jar tests were carried out: first, pH of samples were readjusted to desired pH (pH of 7.5 for $Al_2(SO_4)_3$ and then the varying coagulant concentrations (1000, 2000 and 3000 mg L^{-1} for alum) at room temperature (20^{o}C) were dosed into 1 L of a wastewater sample. The fast mixing for a minute (120 rpm), slow mixing for 30 min. (20rpm) and 1.5 hours sedimentation was applied sequentially in chemical coagulation after that the supernatant was analyzed for COD and turbidity according to Standard Methods.

Granular Activated Carbon (GAC) Adsorption

The supernatant from chemical coagulation process with alum was subjected to adsorption test with GAC. Therefore, the pH of supernatant was first adjusted to 5.5 as suggested by previous study done at the laboratory and 1 L sample volumes having initial COD concentrations of 455, 750, 2520, 5025 and 10970 mg L^{-1} were mixed with constant carbon mass of 10 g (GAC: 0.5-1 mm in size; Merck) for 36 hours period at 80 rpm. The supernatant was filtered through Wattmann 40 filter paper before COD analysis (Fettig et al., 1996).

RESULTS AND DISCUSSION

Alternative I: Pretreatment with lime + ammonia stripping + neutralization

Although the coagulation/flocculation is relatively a simple technique, this treatment only allows to moderate removal of COD and TOC. A few negative situations such as sludge generation and high aluminum concentration was revealed after this process. In the literature, many reports related to chemical treatment of leachate are available. Amokrane et al. (1997) reported that COD in an old landfill leachate was removed between 10% and 25%. Tatsi et al. (2003) obtained COD removal rates of 30% and 45% with using only lime. They advanced removal rates 75% by additioın ferric chloride to lime.

The leachate sample in this study was subjected to lime pretretmant (11 pH). From lime pretreatment, supernatant was coagulated by aluminum sulfate. COD removal rates of 19% and 55% were attained via modifying dosages, respectively. In this study, turbidity removal was found to be 81% for aluminum sulfate (Table 3).

Table 3. Results of experiments with aluminum sulfate ($Al_2(SO_4)_3 18H_2O$)

Jar Test No	Alum Dose (mg L^{-1})	pH	Turbidity (Ntu)		Removal of Turbidity (%)	COD (mg L^{-1})			Removal of COD (%)	
			Raw sample	Effluent from treatment with alum		Raw sample	Effluent from pretreatment with lime	Effluent from treatment with alum	Relative to raw sample	Relative to supernatant from pretreatment with lime
1	1000	7.5	4	0.76	81	23700	19200	10667	55	44
2	2000	7.5	4	1.16	71	23700	19200	13333	44	31
3	3000	7.5	4	2.43	39	23700	19200	16000	33	17

Alternative II: Chemical coagulation + ammonia stripping + granular activated carbon (GAC) adsorption; The obtained results in this study showed that effluent from chemical treatment did not comply with Turkish discharge standards and another polishing step needs to be used. Hence, adsorption by GAC was studied to accommodate discharge standards. Langmuir sorption isotherms, which is one of the most widely used models to describe the equilibrium behavior of adsorption was used to correlate the isotherm data in this study. The Langmuir equation is expressed as in Equation 1:

$$q_{eq} = \frac{Q^o.b.C_{eq}}{1 + b.C_{eq}} \quad (1)$$

Where, Q^o is the maximum amount of the adsorbate per unit weight of adsorbent to form a complete monolayer on the surface bound at final (equilibrium) concentration (C_{eq}) of solute in the solution and b is a constant related to the affinity between the sorbent and sorbate. The lower the value of b expresses the higher the affinity of sorbent. Q^o represents a practical limiting adsorption capacity when the surface is fully covered with adsorbate and assists in the comparison of adsorption performance particularly in cases where the sorbent did not reach its full saturation in experiments. Equation 1 may be written into a linearized form as follows:

$$\frac{C_{eq}}{q_{eq}} = \frac{1}{Q^o b} + \frac{C_{eq}}{Q^o} \quad (2)$$

Q^o and b can be determined from the linear plot of C_{eq}/q_{eq} and C_{eq} (Kestioglu, 1990).

The plots of C_{eq}/q_{eq} versus C_{eq}, respectively were found to be linear with a significantly high regression correlation coefficient (R^2 = 0.9889) and this coefficient is indicate the applicability of the classical maximum adsorption capacity (Q^o) under the experimental conditions was found to be 350 mg COD g^{-1} GAC in this study (Figure 3).

GAC columns (number of 8) were designed using this laboratory data. Each the GAC columns had diameter of 1.5 m, height of 3 m and a total GAC volume of 28 m^3 for following conditions.

Flowrate: Q = 350 m^3 day^{-1}; volumetric loading rate:V_f = 10 m hour^{-1}; Initial COD: C_o = 10970 mg COD L^{-1}; Effluent COD: C_e = 160 mg COD L^{-1}; maximum adsorption capacity after 36 hours of contact: Q^o = 350 mg COD g^{-1} GAC.

Proposed treatment plant flow-schemes

In this study suspended solids, COD and heavy metal in leachate was removed 82%, 19%, 60% by pretreatment with lime, respectively. The landfill leachate could be mixed with municipal wastewater at a rate of 2-5% (Christensen et al. 1992). The using mixture of landfill leachate and municipial wastewater is very useful for treatment costs in developing countries.

Only chemical coagulation process did not sufficiently reduce COD low enough to comply with standards which dictates costly polishing step such as GAC adsorption. If it is a must to comply with strict discharge standards, flow-scheme depicted in Figure 2 could be considered.

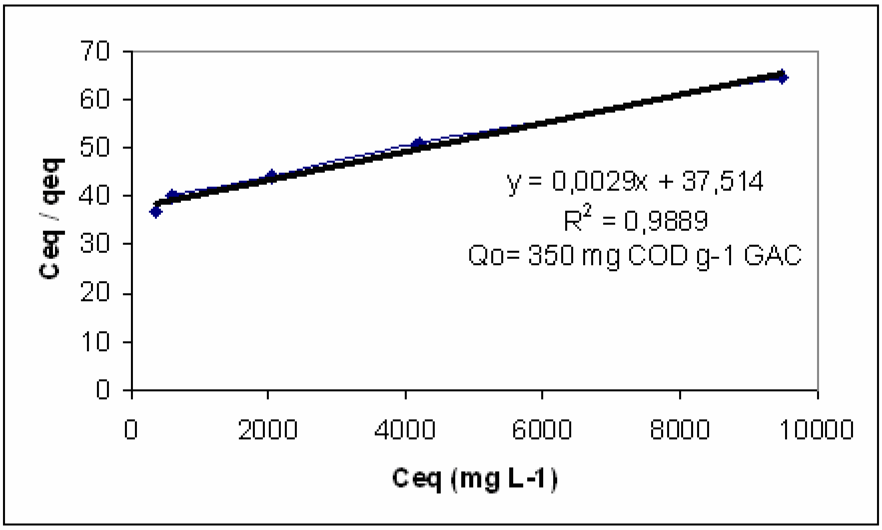

Figure 3. Graphics of Langmuir izoterm

Evaluation of capital and operating costs for each alternative

In cost evaluation, the following items were considered;

- The construction and equipment costs were evaluated in the capital costs
- The operating costs were include such as labor, maintenance, chemical use, sludge disposal and energy consumption

Table 4 presents a comparative cost evaluation for each proposed flow-scheme treating 350 m^3 leachate per day. For capital and operating costs, pretreatment with lime has cheaper than the adsorption process. But, the effluent obtained at the end of the pretreatment was not favourable any discharge. Although, adsorption process produced a good quality effluent that complies with Turkish standards for direct discharging into receiving media (COD = 160 mg L^-) [(Table 20.6), Anonymous 1999)]. It's capital and operating costs were quite high.

Table 4. Comparative evaluation of the treatment alternatives for a daily capacity of 350 m^3

Treatment Alternative	Capital Costs* (USD)	Unit Operating Costs (USD m^{-3})	Annual* Operating Costs (USD $year^{-1}$)	Maintenance* Costs (USD $year^{-1}$)	Annual* Total Costs (USD)	Effluent COD (mg L^{-1})	Compliance with Turkish Discharge Standards
Pretreatment with lime	61,000	5	638,750	8000	646,750	19200	none
Chemical coagulation followed by GAC adsorption	134,000	18	2,299,500 *	20000	2,319,500	160	acceptable for direct discharge into receiving media

- It was assumed that GAC would be completely replaced every two year and a daily steam regeneration with at 120 ^{o} C for each column would be carried out.

CONCLUSIONS

i. The suspended solids, COD and heavy metal in landfill leachate was removed 82%, 19%, 60% through pretreatment with lime, respectively. The mixture of municipal wastewater with pretreated leachate could be proposed for treatment politics of operations. The capital cost and operating cost of pretreatment with lime in the study were found to be \$175 m^{-3} and \$5 m^{-3}, respectively.

ii. Adsorption process followed by chemical coagulation provides an effluent suitable for direct discharge with capital and operating costs of \$383 m^{-3} and \$18 m^{-3}.

Consequently, although alternative I (lime pretreatment and ammonia stripping) is not performed discharge standards; it seems as the more an applicable and cheaper option than adsorption process due to mixing pretreated leachate with municipal wastewater up to 5%. This solution has to be supported by toxicity evaluation. The treatment with adsorption process permits a suitable effluent for directly discharge, but high operating costs are significant issue that must be considered seriously.

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