Volume 22 Issue 38
Originally issued as B3



Global Journal of human-social science: B
Geography, Geo-Sciences, Environmental Science & Disaster Management
Volume 22 Issue 3 Version 1.0 Year 2022
Type: Double Blind Peer Reviewed International Research Journal
Publisher: Global Journals
Online ISSN: 2249-460X & Print ISSN: 0975-587X

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Keywords: leachates, coagulants, jar test, removal efficiency, seasonality.

GJHSS-B Classification: DDC Code: 616.157 LCC Code: RC647.C55



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Abatement of Polluting Effects of Waste Dump Leachates using Different Coagulants

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Abstract- This study assessed the effectiveness of different coagulants for treating leachates before their release into the environment. Three inorganic coagulants (ferric chloride, ferrous sulphate and alum) and one organic coagulant [*Moringer Oleifera* seed (MOS)] were used in a jar test to determine the optimum pH and dosage for the coagulants. Raw and treated leachates were analysed for physiochemical parameters such as pH, chemical oxygen demand, Total solids, Pb and Cr. The optimum pH for ferric chloride, ferrous sulphate, alum and MOS was 7, 7,6 and 10 respectively. While the optimum dosage for each coagulant was 3g/L, 3g/L, 5g/L and 5g/L respectively. The analysis of the raw leachate sample showed that it was highly polluted (Dry season: COD – 3000mg/L, TSS – 2369mg/L, Cr – 0.075mg/L, Pb – 0.25mg/L and Mn – 0.29mg/L; Wet season: COD – 3000mg/L, TSS – 2369mg/L, Cr – 0.075mg/L, Pb – 0.25mg/L and Mn – 0.29mg/L).Coagulants removal efficiency (RE) for COD ranges from 12% to 41% with ferric chloride having the highest removal efficiency. All the coagulants were efficient in reducing the level of heavy metals in the sample leachate. The RE ranges from 55% to 95.6% with MOS having the highest RE of 95.6% for lead. The coagulants showed significant difference (at $P < 0.05$) in their RE for some of the parameters treated. The inorganic coagulants (ferric chloride, ferrous sulphate and alum) showed no significant difference ($P > 0.05$) in the removal of COD, while the organic coagulant (MOS) was significantly different at $P < 0.05$ from the inorganic coagulants. Over all, Alum showed to be a better coagulant than other coagulants in reducing the physiochemical parameters of leachates while MOS is a suitable substitute for alum. It was also observed that there was no significance ($P > 0.05$) in the removal efficiency of the coagulants in both dry and wet seasons. Seasonality has no effect on the effectiveness of the coagulants.

Keywords: leachates, coagulants, jar test, removal efficiency, seasonality.

I. Introduction

The continuous growth in population and industrialization globally has led to increases in solid waste generation and the problem of its management. Solid waste collection and disposal are among the most serious threats to waste management in most cities in developing countries (Donevska *et.al.,* 2006). Solid waste is any material, which is not in liquid form, and has no value to the person who is responsible for it (Zurbrugg, 2003). Babatola (2008) described waste as any material lacking direct value to the user and so must be disposed of.

The poor management of solid wastes constitutes a disaster for human health and leads to environmental degradation (Achankeng, 2003). One of the most important issues of concern in open dump or landfill waste disposal method is the issue of leachate generation and its potential for downgrading water resources systems (Sartaj *et.al.,* 2010). Leachates are defined as the aqueous effluent generated as a consequence of rainwater percolation through wastes, biochemical processes in waste's cells and the inherent water content of wastes themselves (Lee *et.al.,* 2012). The generated leachate can cause significant environmental damage, becoming a major pollution hazard when it comes into contact with the surrounding soil, ground or surface waters. This leachate often contains a high concentration of organic matter and inorganic ions, including ammoniacal nitrogen and heavy metals; posing great treat to human (Zouboulis *et al.,* 2008).

The quality of leachate is affected by factors such as dumpsite age, precipitation, seasonal weather variation, waste type and composition. Treatment methods are highly dependent on leachate characteristics and tolerance of the method against changes in leachate quality such a variable nature along with other factors. The leachate treatments success depends also on the characteristics of the leachate and age of the landfill. Therefore, in order to avoid environmental damage, landfill leachate must be collected and appropriately treated before being discharged into any water body (Oh *et.al*., 2007).

Coagulation is widely used for wastewater treatment. This treatment is efficient to operate and the operating cost is low (Wang *et.al*., 2008). It has many factors that can influence the efficiency, such as the type and dosage of coagulant, pH, mixing speed and time and retention time.The optimization of these factors may influence the efficiency (Wangand Bank, 2007). Coagulation destabilizes the colloidal suspension of the particles with coagulants and then causing the particles to agglomerate with flocculants. After that,it will accelerate separation and thereby makes the effluents clearer (Gnandi *et.al*., 2005).

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There are two kinds of coagulants; inorganic and organic coagulants. Inorganic coagulants (such as Alum, Ferric chloride etc.) are the most commonly used in coagulation treatment of leachate. The use of organic coagulants (M. oleifera seed, Phaseolusvulgaris seed, etc.) is not as common as the inorganic coagulants. The Moringa Oleifera tree grows in tropical and subtropical regions around the world and its seeds have been used in drinking water treatment in small scale in Sudan and India for generations. Coagulation studies are usually carried out using jar test equipment. The jar test has been the typical technique used in wastewater and drinking water industry to improve the addition of coagulant and flocculants (Silver *et.al*., 2004).

This paper seeks to investigate the efficiency of M. oleifera and compare the differences in the removal efficiency of alum, ferrous sulphate and ferric chloride to *M. oleifera* as coagulants in removing physicochemical parameters of leachate. Also to assess the effect of pH on the effectiveness of coagulants in leachate treatment and determine the pollution level of leachate samples by determination of water quality parameters.

II. Materials and Methods

a) Study Area

The study area Saje is located in Abeokuta North Local Government of Abeokuta, the capital of Ogun State, South-West Nigeria. Abeokuta covers an approximate area of about 40.63 km^2. Saje dumpsite lies between latitude 7^0 09' N – 7^0 19' N and longitudes 3^0 29' E – 3^0 41' E (Ufoegbune *et.al.,* 2008).

The Saje dumpsite (figure 1) established in 2006 was formerly a quarry, where mining was done over a long period of time for granites. In order to reclaim the site the state government decided to use the quarry as dumpsite. The dumpsite is the only major dumpsite used in Abeokuta metropolis and is about 4 ha in area. Saje area was formally an outskirt of Abeokuta town but due to increased population of the metropolis, houses have encroached the site of the dump site (Badejo *et.al.,* 2013).

The location coordinate of the dumpsite was obtained with a hand held Global Positioning System (GPS, Garmin MAP 76CSx model made in Taipei County, Taiwan) with position accuracy of less than 3m. The choice of the sampling points within the dumpsite was considered using the following criteria: location, accessibility and availability of leachate.

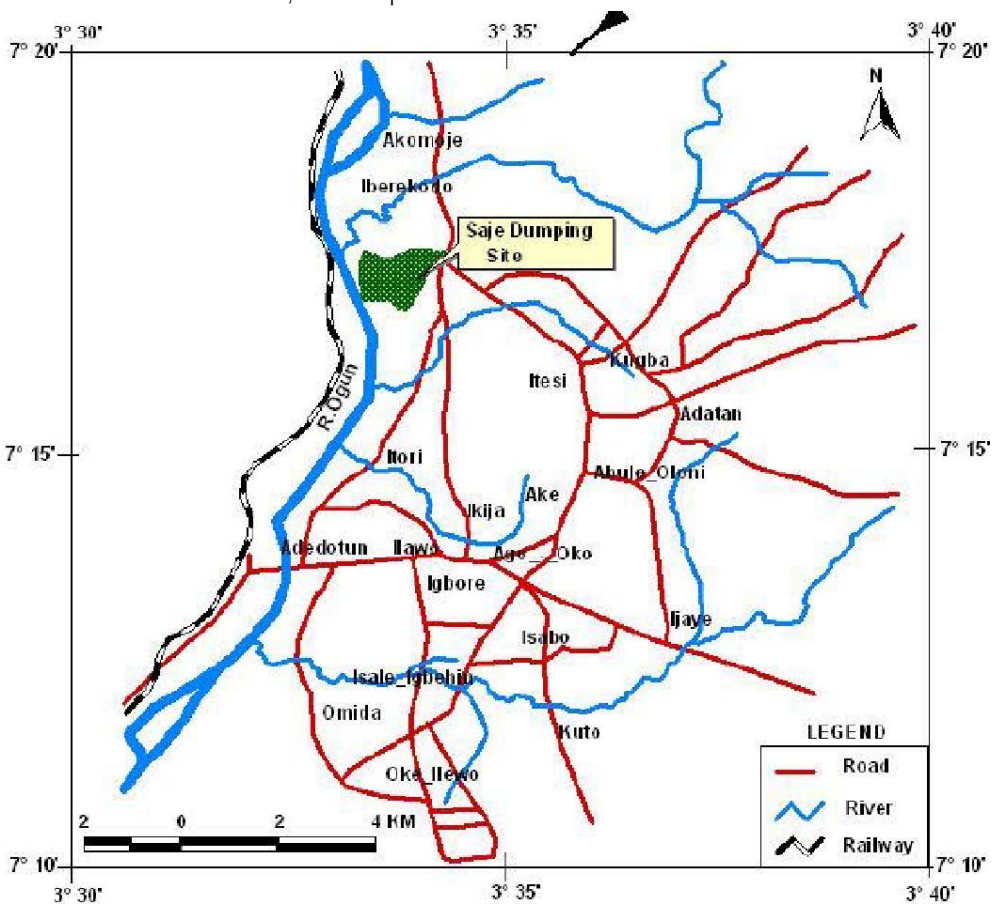

Figure 1: Saje Dumpsite Location within Abeokuta City, Ogun State, Nigeria.

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b) Sampling Procedures

Leachate: Leachate samples were collected from Saje dump site (figure 1) in Abeokuta, Ogun State during wet and dry seasons using standard methods. Samples were collected in 15 L plastic containers, transported to the laboratory and stored in a refrigerator at 4°C. The samples were removed from the refrigerator and left under room temperature for at least 2 hours. Then, the samples were thoroughly stirred to agitate settled solids before any tests were conducted on the leachate samples.



Figure 2: Leachate collection point from Saje dumpsite

c) Reagents

In this study, Ferric Chloride ($FeCl_3.6H_2O$), Ferrous Sulphate ($Fe(SO_4).7H_2O$) and Aluminum Chloride ($Al_2(SO_4)_3.18H_2O$) were used as inorganic coagulants. *Moringa oleifera* Seed (MOS) was used as organic coagulant. MOS 10% Stock solution was prepared daily by dissolving 30g of MOS powder into 270ml of distilled water, and the solution was well mixed.

d) Analysis Techniques

The physical and chemical parameters were determined using APHA Standard Methods (2005) for testing water and waste water. pH was assessed by glass electrode method with a calibrated pH meter, while temperature EC and TDS was determined using HM Digital Meter COM-100. Total alkalinity, total hardness, Acidity, chloride, were determined by titrimetric method. A total suspended solid was determined by gravimetric method. Chemical oxygen demand (COD) was determined by open reflux method. Nitrate Phosphate and Sulphate were measured by UV-Visible spectrophotometer. The heavy metal analysis was carried out using Atomic Absorption Spectrophotometer (AAS) Model 210 VGP of the Buck Scientific AAS series.

e) Experimental Procedure

Chemical coagulation was performed using beakers and stirrer as Jar test apparatus. The experimental process consisted of three subsequent stages: initial rapid mixing at 160 rpm for 10 min, followed by slow mixing for 20 min at 30 rpm, the final settling time for 1 h.

First, the optimum pH was determined by varying the pH of the sample using HCl and NaOH at constant coagulant concentration. The pH with the highest removal efficiency was the optimum pH.

About 2L beakers of equal volume were used to examine the different coagulants at their respective optimum pH. A known mass of (1g, 2g, 3g, 4g and 5g) of each coagulants was added to a jar containing 1liter of leachate samples at optimum pH using the jar test procedure. To determine the efficiency of coagulant dose, the supernatant was withdrawn by using a pipette from a point about 2 cm below the top of liquid level of the beaker and the supernatant was assessed for TSS, COD, Mn, Pb and Cr.

f) Data Analysis

Data collected were evaluated for descriptive and inferential statistics using the Statistical Package for Social Sciences (SPSS) for windows version 20.The removal efficiency (RE) of the coagulants was determined for each parameter by using the equation:

$$\text{RE}\ (\%) = \frac{Ci - Cf}{Ci} \ x\ 100 \frac{Ci - Cf}{Ci} \ x\ 100$$

Where, C_i and C_t are the initial and final concentrations of the parameters.

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III. Results and Discussion

a) Characteristics of Landfill Leacahte

The results of the physiochemical analysis of the untreated leachate samples from the dumpsite during dry and wet seasons are presented in Table 1.

The Chemical Oxygen Demand (COD), Total Suspended Solids (TSS), Total Dissolved Solid (TDS), Chloride, Chromium, Lead and Manganese of the untreated leachate samples from the dumpsites exceeded the limiting values recommended by the WHO and the FMENV. The values of all other parameters were within the allowable limits as specified. The high level of Pb and Mn is due to the dumping of metals such as cans, used batteries, iron etc in the dumpsite.

Table 1: Characteristic of raw leachate sample for wet and dry seasons

Parameters	Dry (mg/L)	Wet (mg/L)	T value
*pH	7.50±0.10	7.70±0.20	0.011
Alkalinity	47.00±1.00	49.00±1.00	0.007
COD	2900±101	3000.0±100	0.01
Acidity	50.00±10.10	47.00±3.0	0.006
Hardness	2240±201	2280.0±102	0.01
**EC	5790±120	8740.0±90.0	0.02
TDS	3110±110	4720.0±96.0	0.004
TSS	2333.0±120.00	2333.0±20.0	0.001
Chloride	870.0±65.00	910.0±202	0.01
Nitrate	0.73±0.00	0.31±0.00	0.01
Phosphate	0.49±0.00	0.32±0.00	0.012
Sulphate	204.51±2.52	174.84±10.0	0.01
Chromium	0.07±0.01	0.08±0.01	0.013
Lead	0.25±0.0	0.25±0.03	0.013
Manganese	0.29±0.02	0.30±0.02	0.014

**No units **µS/cm*

b) Coagulants optimum pH

Results for the optimum pH obtained from the coagulation of leachate samples using Ferric Chloride, Ferrous Sulphate, Aluminum Chloride and MOS at varying pH values (4,6,7,8, and 10) to evaluate COD and TSS concentrations in the samples are shown in Figure 3 and 4. All the coagulants were kept at 2g/L in all the runs. The pH with the highest removal efficiency (ER) was taken as the optimum pH for the coagulant. It was noticed that all the coagulants gave different results at different pH. $FeCl_3$ and $FeSO_4$ had optimum pH of 7 and Alum had optimum pH of 6, while MOS had its optimum pH at a pH value of 10.

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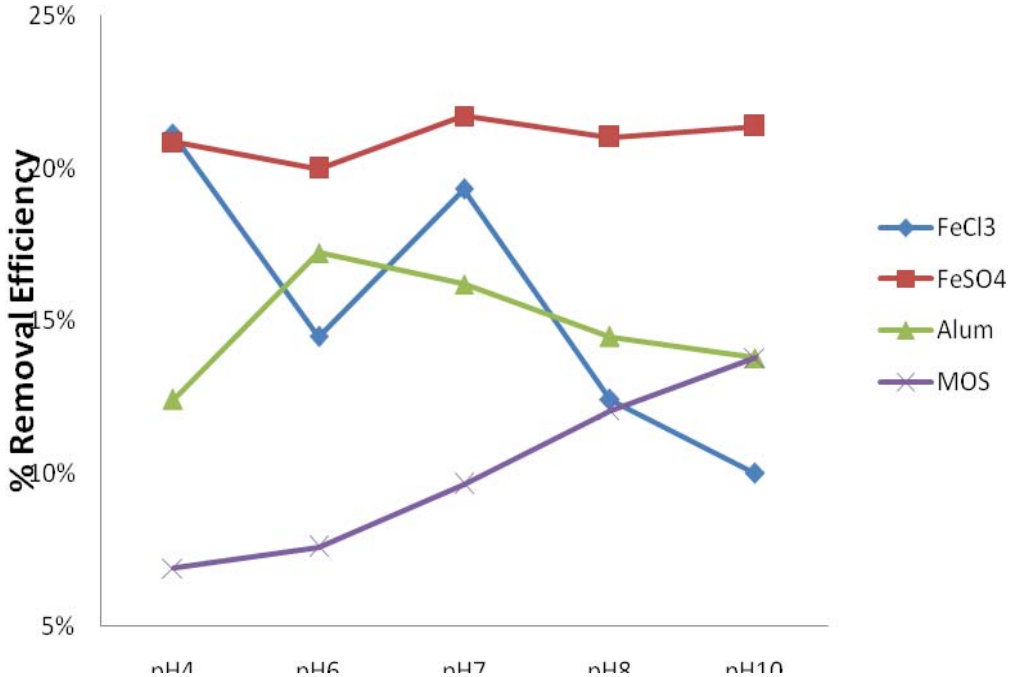

Figure 3: Effects of varying pH at constant coagulant dosage on COD

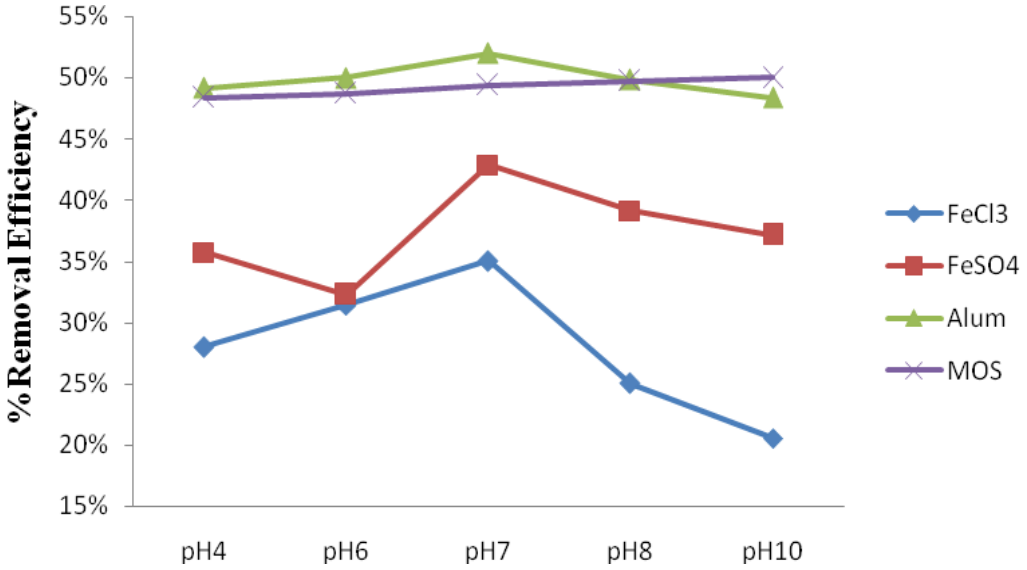

Figure 4: Effects of varying pH at constant coagulant dosage on TSS

c) Effects of Different Coagulant Concentrations in Coagulation Treatment

To observe the effect of coagulant dose, the experimental runs were conducted at different doses (1, 2, 3, 4 and 5 g/L). The percentage removal efficiency at each dose was compared. Depending on the coagulants, the optimal dose varied with the various coagulants used.

$FeCl_3$ removal efficiency for heavy metals ranged from 55% to 85% (Figure 5), this is in line with the reported work of Lee *et.al*. (2012) where $FeCl_3$ was reported to remove 75% of Pb. Amuda and Alade (2006) also gave a report in this range. $FeCl_3$ removed Cr better than Pb and Mn. $FeCl_3$ was not as efficient in removing COD, the value ranged from 19% to 40% (Figure 6). Other studies also reported low RE of $FeCl_3$ for COD (Ibrahim *et.al*., 2012; Lee *et.al.,* 2012). The optimum dosage for $FeCl_3$was determined to be 3 g/L, the RE dropped beyond this dosage.

$FeSO_4$ removal efficiency for heavy metals ranged from 65% to 85% as shown in Figure 7. $FeSO_4$ also removed more of Cr when compared to Pb and Mn, following the trend of $FeCl_3$. It was also not as efficient in removing COD, the value ranges from 21% to 37% (Figure 8) this was in accordance with the work of Ibrahim *et.al*. (2012). $FeSO_4$also had optimum dosage of 3 g/L. Additional concentration above the optimum dosage reduced the efficiency of the coagulant.

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Alum had a higher RE for heavy metals compared to $FeCl_3$ and $FeSO_4$. Its values ranged from 72% - 94.28%. Like in other Coagulants, Cr has the highest RE of 94.28%. This was closely followed by Pb (92.8%) and Mn (87.9%) as presented in Figure 9. The maximum COD removal of 41.72% (Figure 10) resulted at 5 g/L coagulant dose. The COD removal increased with alum dosage increase. Zazouli and Yousefi (2008), Bila *et.al*. (2005) also reported RE of Alum for heavy metals in the range of 71% - 96% and COD in the range of 27% - 40% in their reports. Meanwhile, Trebouet *et.al*. (2001) reported a much lower maximum RE of 66% for heavy metals.

MOS had the best range of RE for heavy metals of all the four coagulants with a minimum of 72.4% and maximum of 95.6% (Figure 11). It removed more of Pb, than Mn and Cr had the least RE. Ravikumar and Sheeja, (2013) reported a 93% RE for Pb and 70% RE for Cr in their work. Both Alum and MOS increased there RE for heavy metals with increase in concentration. Figure 12 showed that MOS treated samples had increased COD concentrations, giving a negative RE. This is similar to the report in previous studies (Arnoldsson and Bergman, 2007).

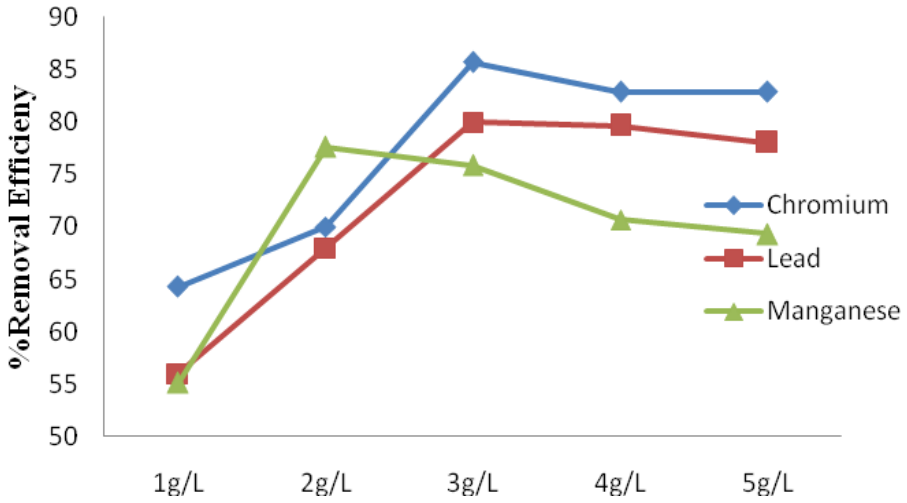

Figure 5: $FeCl_3$ percentage removal efficiency for Cr, Pb and Mn

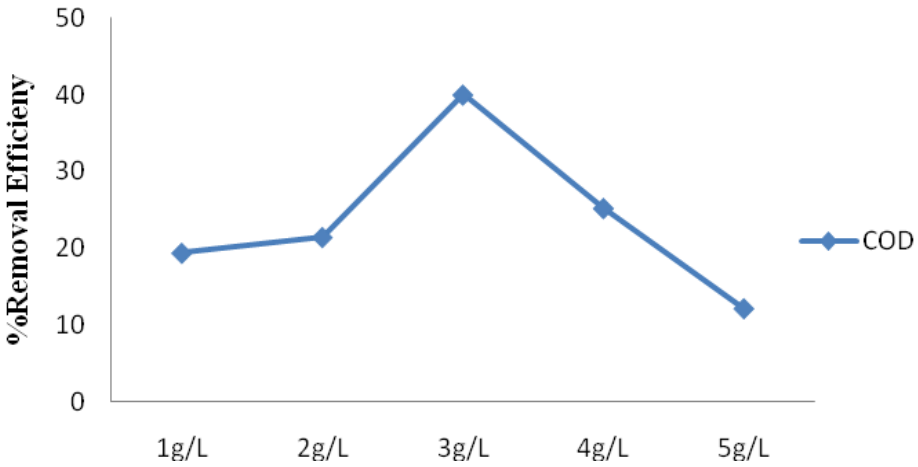

Figure 6: $FeCl_3$ percentage removal for COD

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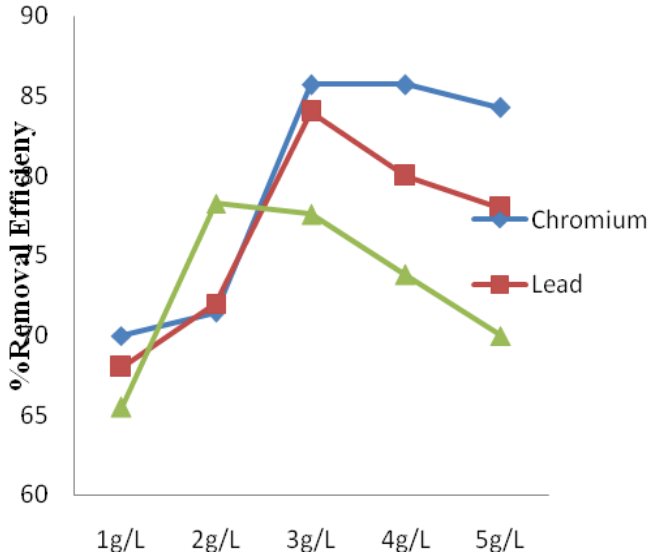

Figure 7: $FeSO_4$ percentage removal efficiency for Cr, Pb and Mn

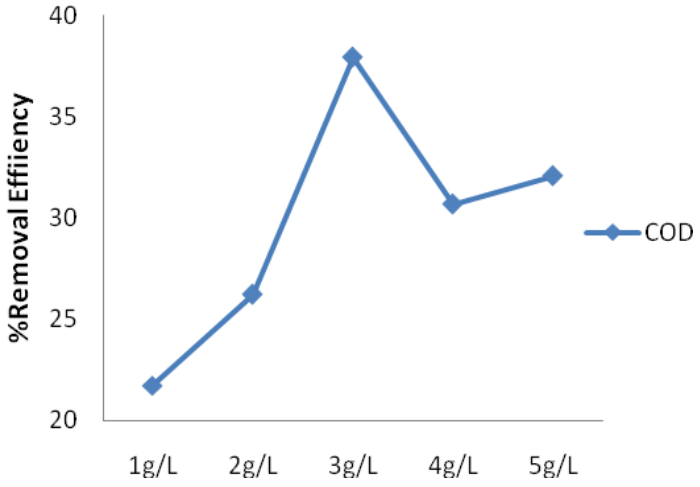

Figure 8: $FeSO_4$ percentage removal efficiency for COD

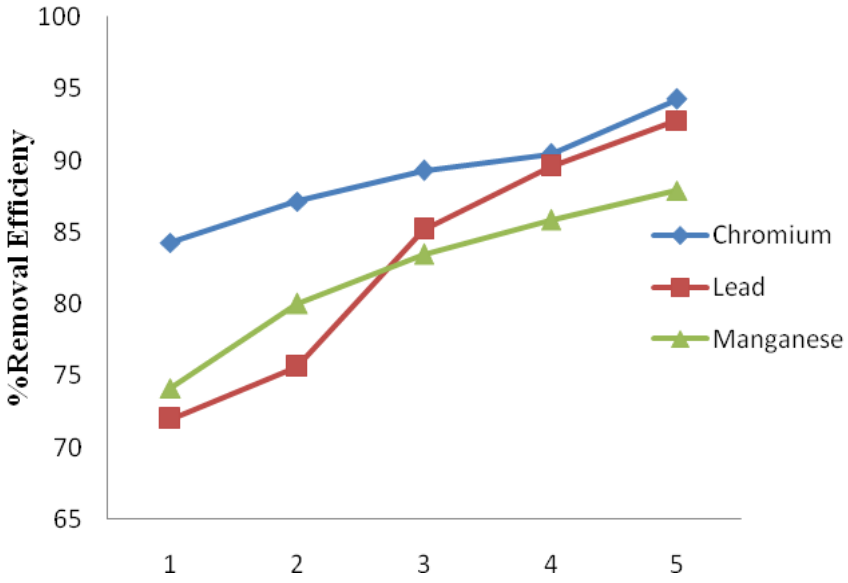

Figure 9: Alum percentage removal efficiency for Cr, Pb and Mn

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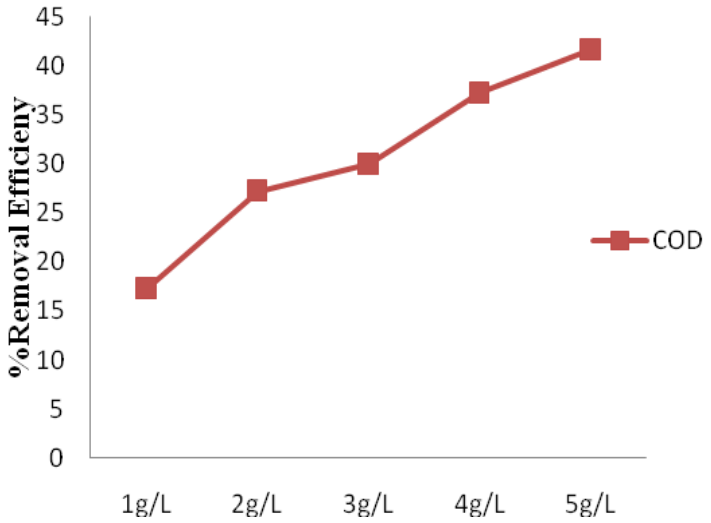

Figure 10: Alum percentage removal efficiency for COD

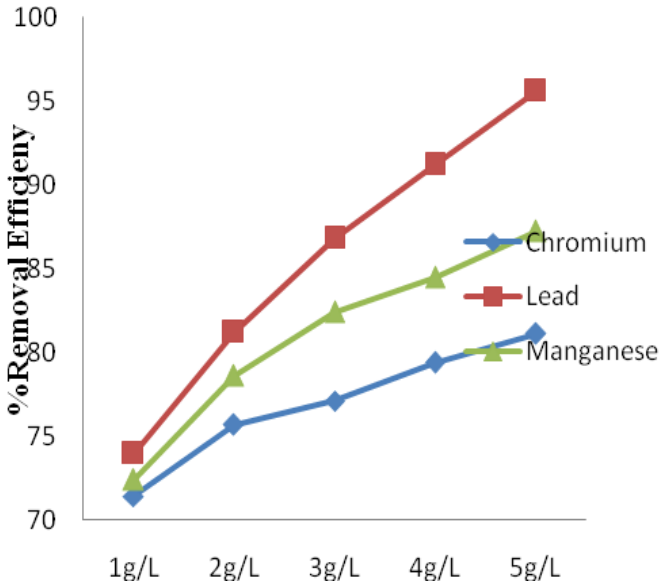

Figure 11: MOS percentage removal efficiency for Cr, Pb and Mn

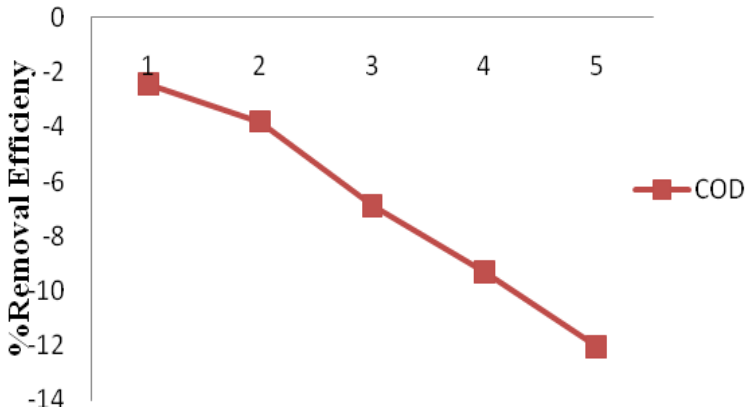

Figure 12: MOS percentage removal efficiency for COD

Alum exhibited a good performance in the sample parameters compare to other coagulants. Table 2 shows that the coagulants were able to reduce heavy metals from the leachates samples to level below standard limits but COD and TSS still had values higher than the recommended standards.

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Table 2: Summary of comparison of coagulants optimum dosage parameters

Parameters	Raw	$FeCl_3$(3g/L)	FeSO4(3g/L)	ALUM(5g/L)	MOS(5g/L)	WHO Standard	FMENV Limit For Discharge To The Environment
COD (mg/L)	3000	1770	1875	1730	0	60.9	60.9
TSS (mg/L)	2369	207	1637	368	1047.5	25	25
Chromium (mg/L)	0.0750	0.0105	0.0105	0.0045	0.0133	0.0500	0.0500
Lead (mg/L)	0.2500	0.0490	0.0395	0.0175	0.0110	0.0500	0.0500
Manganese (mg/L)	0.2950	0.0705	0.0645	0.0355	0.0365	0.0500	-

Over all, Alum was a better coagulant than the three other coagulants in reducing the physical and chemical parameters of leachates.

d) Seasonal Variations of Leachate Concentration

The test was carried out both in the dry and wet seasons to determine effect of season on the efficiency of coagulants. Table 4 and 5 is a summary of the physical and chemical properties of the raw and treated leachate samples for dry and wet seasons.

It was shown that the physical and chemical properties of the leachates are higher in the wet season than in the dry season. This can be attributed to the fact that rainfall is a crucial factor in the formation of leachate and the characteristic of the leachate.

Season has no effect on the efficiency of coagulants. The trend of the removal efficiency of each coagulants tested in dry season is similar to that of the wet season.

Table 4: Summary of Physical and Chemical Parameters of Raw Leachates and Treated Leachate in the Dry Season

	Raw Leachate	After treatment $FeCl_3$	After treatment $FeSO_4$	After treatment Alum	After treatment MOS
COD (mg/L)	2900.0 ± 100.50^{b}	2216.0 ± 274.32^{a}	2038.0 ± 177.68^{a}	2010.0 ± 274.32^{a}	3100.0 ± 268.23^{b}
TSS (mg/L)	2333.0 ± 120.00^{c}	1037.9 ± 295.90^{a}	1726.5 ± 383.22^{b}	717.56 ± 295.90^{a}	1246.5 ± 159.15^{ab}
Cr (mg/L)	0.07 ± 0.01^{c}	0.02 ± 0.00^{b}	0.01 ± 0.01^{ab}	0.01 ± 0.00^{a}	0.02 ± 0.00^{b}
Pb (mg/L)	0.25 ± 0.03^{c}	0.07 ± 0.02^{b}	0.06 ± 0.02^{ab}	0.04 ± 0.02^{a}	0.04 ± 0.02^{a}
Mn (mg/L)	0.29 ± 0.02^{c}	0.09 ± 0.02^{b}	0.08 ± 0.02^{b}	0.05 ± 0.02^{a}	0.06 ± 0.02^{a}

Table 5: Summary of Physical and Chemical Parameters of Raw Leachates and Treated Leachate in the Wet Season

	Raw Leachate	After treatment $FeCl_3$	After treatment $FeSO_4$	After treatment Alum	After treatment MOS
COD (mg/L)	3000.0 ± 100^{b}	2350.0 ± 288.57^{a}	2170.0 ± 163.25^{a}	2122.0 ± 288.57^{a}	3270.00 ± 279.52^{b}
TSS (mg/L)	2333.0 ± 20.0^{c}	1074.2 ± 295.98^{a}	1591.0 ± 136.38^{b}	717.6 ± 295.98^{a}	1246.6 ± 159.21^{ab}
Cr (mg/L)	0.08 ± 0.01^{c}	0.02 ± 0.00^{b}	0.02 ± 0.01^{ab}	0.01 ± 0.00^{a}	0.02 ± 0.00^{b}
Pb (mg/L)	0.25 ± 0.03^{c}	0.07 ± 0.02^{b}	0.06 ± 0.02^{ab}	0.04 ± 0.02^{a}	0.04 ± 0.02^{a}
Mn (mg/L)	0.30 ± 0.02^{c}	0.09 ± 0.02^{b}	0.08 ± 0.02^{b}	0.05 ± 0.02^{a}	0.06 ± 0.02^{a}

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IV. Conclusion

The application of coagulation treatment for raw leachate collected from Saje dumpsite showed the leachate was characterized by low pH and high concentration of pollutants; especially that of organic matter as observed in the COD level and high level of heavy metals which are all above the WHO and the FMEnv limit for waste water. The study showed that the leachate from the dumpsite is polluted and there is need for it to be treated before it is released into environment.

The study showed that coagulation treatment is efficient in ameliorating the polluting potential of dumpsite leachates. All the four coagulants; ferric chloride, ferrous sulphate, alum and MOS were able to reduce the heavy metals in the leachate by over 55% and MOS removing as high as 95.6%. MOS was better than the other coagulants in terms of removal efficiency for heavy metal. The coagulants were not as effective against COD, with alum giving the highest removal efficiency of 41.7% and MOS increased the COD concentration. None of the coagulants was able to bring the COD level down to below the FMEnv standard limit.

This study also revealed pH as an important factor in coagulation. It was established that each coagulant has the pH at which it works best; to remove contaminants. This pH isreferred to as the optimum pH. In this study the optimum pH for Ferric chloride and ferrous sulphate was 7.0, Alum was 6.0 and MOS was 10.0.

This study had determined the optimum dosage of each coagulant to get the best use of them. It was observed that the optimum dosage for ferric chloride, ferrous sulphate, alum and MOS were 3.0g/L, 3.0g/L, 5.0g/L and 5.0g/L respectively. From the results Alum was the best coagulant for treating leachates, closely followed by ferric chloride, MOS and ferrous sulphate in that order.

This study has shown little or no seasonal variation in the concentration of leachate. The season did not have significant effect on the efficiency of the coagulants

Moringa Oleifera showed good coagulating properties, and has many advantages compared to aluminium sulphate. It did not affect the pH, alkalinity or conductivity of the water, and it can be produced locally at low cost. *Moringa oleifera* is an environmentally-friendly natural coagulant that can be used to replace alum and other inorganic coagulants particularly in treating drinking water. It is a method that certainly can be considered as a good, sustainable and cheap solution for smaller waterworks, if the supply of Moringa seeds can be guaranteed.

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