REMEDIATION OF DUMPSITE LEACHATE CONTAMINANTS BY COAGULATION AND COMPLEXATION

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ABSTRACT

Leachate is a major threat to groundwater quality and ecosystem. The present study adopted four selected coagulants (inorganic: $Al_2(SO_4)_3$ [Alum], $FeCl_3$, $FeSO_4$; biological: *Moringa oleifera*) and a complexing agent (ethylenediaminetetraacetic acid EDTA) to remediate leachates collected from two dumpsites in Abeokuta. The leachate samples were subjected to chemical and heavy metal analyses using the American Public Health Association (APHA) standard method. The data obtained were evaluated for simple descriptive statistics (mean and standard deviation) using SPSS for Windows. The results indicated that except for Fe and Ni, metal values in both raw and treated leachates were generally higher than the permissible standard of the National Environment Management Authority (NEMA). The study revealed that despite treatment, the values of metals in treated leachates were still issues of environmental concerns. The reduction data revealed substantial removal of EC and TDS (100%), Cl^- (87-95 %) and SO_4^{2-} (60-84%) in leachates. The present study also revealed that each coagulant has varied affinity for removal of different parameters in the leachate. Among the coagulants, *Moringa oleifera* showed the highest % removal efficiency for Cd (57-63%) and Ni (57-98%), while EDTA showed the lowest removing ability for Cd (2-6%). All the remediating agents effectively reduced the chemical contaminants in leachate samples to appreciate levels.

Keywords: Coagulants, EDTA, Leachate, dumpsite, chemical parameters, remediation

INTRODUCTION

Leachate is the liquid produced from decomposition of landfill waste and biodegradation process [1]. This is usually aided by humidity, water contents of residual organic matter in compartment layers and controlled factors including environmental conditions, landfill operational management and decomposition dynamism within the compartments [2]. The factors, which determine leachate characteristics, vary with respect to its composition, volume and biodegradable matter against time [3, 4]. The characteristics of leachate are determined by the dumpsite age; for example, young (< 5 years), intermediate (5-10 years) and stabilized (> 10 years) [5, 6]. Inappropriate handling of landfill influences ecological risks due to leachate generation and discharge that percolate the sub-soil with resultant adverse effects on ground and surface waters [7, 8]. Leachate from dumpsites and landfills had re-

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portedly streamed through the paddy fields onto the nearby water sources where it caused surface water pollution [9]. The infiltration of leachate into the water table and mixing with groundwater source may result into formation of pollution plume, which may spread in the flowing groundwater direction [10]. The presence of polluting and toxic substances in leachate may initiate numerous ecotoxicological and health ill-effects [11, 12]. The four major constituents of leachate are dissolved organic matter, inorganic macro-components, heavy metals and xenobiotic organic compounds [13]. The human health effects of the toxic substances in leachate had been assessed by Swati *et al.,* [14]. It is therefore important to handle leachate management properly to avoid pollution of water and soils [15]. The present weak regulation, low technology know-how and quantity of leachate generated daily from dumpsites in the developing countries are major contributors to ecological toxicity [16, 17].

The different remediation methods of leachate/effluents for pollutants are coagulation/ flocculation, precipitation processes, ion-exchange, adsorption, membrane filtration, sedimentation and electrochemical [18-21]. The appropriate method for remediation depends on the characteristics of leachate; however, coagulation or flocculation process is one of the most efficient techniques [22]. These remediating processes may be classified broadly into biological and physical/chemical technologies; but combination of the two methods may be adopted depending on the scales and goals [23].

Coagulation or flocculation is a physico-chemical process efficient for removing suspended and colloidal particles [24]. The study on comparative efficiency of coagulants revealed that a number of coagulants may lead to better results [22]. In previous studies, the effectiveness of chemical coagulants such as aluminum sulfate, ferrous sulfate, aluminum chloride and ferric chloride has been compared [25, 26]. The use of *Moringa oleifera* seed as a natural coagulating agent for leachate treatment is becoming more popular [12]. Several studies have investigated the efficient applications of *Moringa oleífera* seed for leachate treatment [27-30]. EDTA has also been proved to be an effective method of removing heavy metals in polluted samples due to its strong complexing ability [31]. However, much has not been done on the removal efficiency of metals in leachate using EDTA. The few studies on the use of EDTA for metal removal were conducted on soils [31-33]. The objective of this study is to remediate dumpsite leachate using different coagulants and EDTA.

MATERIALS AND METHODS

Leachate collection and analysis

Leachate samples were collected in September 2017 from two major dumpsites in Abeokuta. Samples were collected in 25 L plastic kegs and transported to the Laboratory of the Department of Environmental Management and Toxicology, Federal University of Agriculture, Abeokuta. Leachate samples were refrigerated at 4ºC in the dark in order to minimize leachate decomposition due to microbial activity [12, 34]. Raw leachate samples were determined for important chemical parameters using standard procedure [35]. The leachate samples were treated with four coagulants and EDTA prepared in the varying concentration doses of 1000, 3000 and 5000 mg/L. The chemical parameters determined in leachates were pH, electrical conductivity (EC), total dissolved solid (TDS), chloride, nitrate, sulphate, phosphate and heavy metals (Cd, Cr, Fe, Pb, Mn, Ni). pH, EC and TDS were determined electrochemically using the combined pH/TDS and EC tester (HI98130, Hanna). The water soluble ions (Cl^-, NO_3^-, SO_4^{2-}, PO_4^{3-}) were analyzed using spectrophotometric method (Hach DR/4000, UK), while metals were analyzed using the Atomic Absorption Spectrophotometry (AAS, Bulk Scientific, 210/211VGp, CT, USA). Before the determination of heavy metals, leachate samples were digested with concentrated HNO_3 by measuring 100 mL of leachate into a clean prewashed conical flask followed by addition of 10 mL concentrated HNO_3 (Sigma-Aldrich Chemie GmbH, Darmstadt, Germany) and then heated on a hotplate for 30 minutes. The digested samples were filtered using Whatman filter paper of 1.0 µm pore size. The digested extract was sent to the Central Laboratory of University of Ibadan for AAS (AAS, Bulk Scientific, 210/211VGp, CT, USA) analysis. The AAS instrument was calibrated with series of standard solutions (Cd, Cr, Fe, Pb, Mn, Ni) prepared in the concentration range of 0–6000 mg L^{-1}, to obtain the calibration curves. Chemical reagents used for metal analysis were of Analytical Grade (Sigma-Aldrich Chemie GmbH, Darmstadt, Germany). Blank samples were also analyzed to cancel the matrix effects of extracting reagents and to calculate the limit of detection of instruments as 3 x standard deviation [36].

Preparation of coagulating/EDTA treatments

The *Moringa oleifera* seeds were air-dried, shelled and pulverized with electric blender [12]. The dosage treatments of 1000, 3000 and 5000 mg/L were prepared by weighing exactly 2, 6 and 10 g of *Moringa oleifera* into 2 L of raw leachate samples. Similarly, other treatment doses for Alum, $FeCl_3$, $FeSO_4$ and EDTA were prepared in the varying concentrations of 1000, 3000 and 5000 mg/L. Each treatment was replicated six times. The experimental setups were stirred for 1 hour and left for 72 hours. After treatment, the mixture was filtered with Whatman filter paper of pore size 1.0 µm [37]. The treated leachates were then subjected to chemical analysis.

Data analysis

Data collected were evaluated for simple descriptive statistics (Mean and Standard deviation) using SPSS for Window version 22.0. The percent reduction of leachate parameters were calculated as:

$$\% \text{ Reduction} = \frac{Xi - Xf}{Xi} \times 100 \text{ [38]}$$

Where, X_i and X_f refer to the initial and final values of measured parameters, respectively.

RESULTS AND DISCUSSION

Chemical parameters of leachates before and after treatment

Results of the chemical properties of both the raw and treated leachate samples are presented in Table 1. The pH of the raw leachate was 8.15±0.49 indicating an alkaline medium. This pH value was similar to the pH range of 7.77 - 8.19 reported by Ojoawo *et al.,* [39] in leachate samples collected from dumpsites in Ogbomosho, Oyo state. The introduction of inorganic coagulants and EDTA reduced the leachate pH to acid medium. However, *Moringa oleifera* maintained the alkaline environment in the treated leachate, while Alum, $FeCl_3$ and $FeSO_4$ changed the pH to acidic medium. As the concentrations of EDTA and *Moringa oleifera* increased from 1000 to 5000 mg/L, the pH decreased; while increasing in concentrations of Alum, $FeCl_3$ and $FeSO_4$ increased the pH of the dumpsite leachate.

The EC (25350±11101 µS/cm) and TDS (12990±5954 mg/L) concentrations of the raw leachate were higher than the permissible standards of 1000 µS/cm [40] and 2000 mg/L [41], respectively. However, the study of Aiyesanmi and Imosi [42] has measured higher concentrations of EC (13719-64344 µS/cm) and TDS (6834-3.2192 mg/L) in dumpsite leachates sampled in Benin City. EC and TDS are environmental issues of concern in terms of soil and groundwater pollution with dissolved salts. The four coagulants and EDTA were able to reduce EC and TDS to levels that are within the permissible limits indicating the effectiveness of the remediating agents.

The concentrations of Cl^-, PO_4^{3-} and SO_4^{2-} in raw leachates were generally lower than the permissible standards of the Federal Environmental Protection Agency [41]. NO_3^- was measured below detection limits. The concentrations of Cl^-, PO_4^{3-} and SO_4^{2-} measured in raw leachates were lower than the values previously reported by Ojoawo *et al.,* [39]. After remediation, Cl^-, PO_4^{3-} and SO_4^{2-} reduced by at least 40% indicating the promising potentials of used coagulants and EDTA. The concentrations of PO_4^{3-} in the treated leachate at

TABLE 1

Chemical properties of leachates before and after treatment with varying concentrations of coagulants

		pH	EC (µS/cm)	TDS (mg/L)	Cl^- (mg/L)	NO_3^- (mg/L)	PO_4^{3-} (mg/L)	SO_4^{2-} (mg/L)
Raw Leachate		8.15±0.49	25350±11101	12990±5954	33.89±20.81	<0.01	19.69±7.24	84.50±16.26
Moringa oleifera	1000 mg/L	7.75±0.01	121.15±0.28	27.91±0.16	0.14±0.04	<0.01	5.25±1.77	39.45±1.62
	3000 mg/L	8.68±0.01	117.35±0.28	25.89±0.02	0.02±0.01	<0.01	4.47±1.06	32.72±0.31
	5000 mg/L	8.34±0.35	120.43±1.02	26.68±0.02	0.02±0.01	<0.01	3.59±1.87	34.55±1.10
EDTA	1000 mg/L	4.08±0.02	144.65±0.78	5.92±0.01	0.12±0.01	<0.01	2.87±0.47	24.52±8.87
	3000 mg/L	4.06±0.00	152.30±0.28	3.93±0.02	0.13±0.04	<0.01	3.13±0.71	16.22±3.07
	5000 mg/L	4.59±0.01	143.65±0.07	4.56±0.01	0.05±0.06	<0.01	2.55±0.74	17.52±5.26
$Al_2(SO_4)_3$	1000 mg/L	6.67±0.02	115.85±0.57	1.41±0.01	0.13±0.02	<0.01	4.83±1.59	23.28±3.93
	3000 mg/L	5.87±0.00	123.33±0.18	1.94±0.01	0.16±0.02	<0.01	5.40±0.56	23.51±2.88
	5000 mg/L	5.62±0.01	123.53±0.25	2.83±0.02	0.11±0.01	<0.01	5.00±0.51	23.56±3.02
$FeCl_3$	1000 mg/L	3.89±0.01	145.03±0.52	7.93±0.01	0.14±0.01	<0.01	4.66±0.34	16.38±1.46
	3000 mg/L	3.83±0.02	150.50±0.99	14.79±0.10	0.09±0.01	<0.01	4.53±1.00	18.43±2.72
	5000 mg/L	3.56±0.03	148.40±0.78	14.65±0.30	0.09±0.08	<0.01	4.31±1.15	16.45±1.28
$FeSO_4$	1000 mg/L	6.03±0.02	123.10±0.52	3.56±0.01	0.12±0.01	<0.01	3.59±0.92	29.90±2.68
	3000 mg/L	6.47±0.01	116.10±0.57	5.06±0.01	0.20±0.02	<0.01	2.63±1.03	32.45±1.27
	5000 mg/L	4.09±0.02	147.35±0.78	8.73±0.02	0.21±0.02	<0.01	2.85±0.25	29.63±0.20
	FEPA [41]	6-9	1000*	2000	100	20	50	100

**WHO [40]*

varying dosage concentrations of remediating agents were still high to cause nutrient enrichment of water bodies. In natural water, PO_4^{3-} concentration is normally measured at concentration less than 0.03 mg/L [43]. Generally, phosphate pollution is one of the major threats to water quality in Nigeria [44].

The metal concentrations of raw and treated leachates are presented in Table 2. All the metals (except Fe and Ni) in raw leachate samples exceeded the National Environment Management Authority [45] permissible limits. However, the Fe content in raw leachates was higher than the FEPA [41] allowable limit of 0.05 mg/L. High level of Fe in leachate may lead to dark brown colour associated with oxidation of ferrous to ferric form, and formation of ferric hydroxide colloids and complexes with fulvic/humic substances [9]. The coagulants and EDTA were able to remediate significant amount of metals from the raw leachate samples. Even though, none of the remediating agents was able to reduce the metal concentrations to zero level; but has shown varying degrees of removal efficiency. After remediation, metal contents (e.g. Cr, Pb and Mn) in some of the treated leachate samples still exhibited concentrations greater than the permissible limits of the National Environment Management Authority [45]. Comparing these metal concentrations in the treated leachates with FEPA [41] permissible standards revealed levels that can result into contamination of ground and surface waters, and also bio-accumulate in plants and aquatic organisms [46].

Percent removal of chemical parameters

Figure 1 shows the effects of coagulants and EDTA on leachate's chemical parameters. EC and TDS were totally remediated by all the coagulants and EDTA at differing concentrations of treatments. In the study of Raghab *et al.,* [23], TDS removal by adsorbents only decreased by one-quarter. $FeCl_3$ and EDTA reduced pH of raw leachates by at least 50%. *Moringa oleifera* was also able to reduce the pH at treatment dose of 1000 mg/L. As the treatment dose of *Moringa oleifera* increased from 3000 to 5000 mg/L, the pH also increased by 7 and 2%, respectively. All the remediating agents performed excellently in removing Cl^- by 87-95%. Among the soluble ionic species, PO_4^{3-} was least removed by all the remediating agents.

TABLE 2
Metal composition of leachate before and after treatment with varying concentrations of coagulants

mg/L		Cd (mg/L)	Cr (mg/L)	Fe (mg/L)	Pb (mg/L)	Mn (mg/L)	Ni (mg/L)
Raw Leachate		0.59±0.08	1.60±1.12	2.96±0.32	0.31±0.08	2.21±0.59	0.26±0.01
Moringa	1000 mg/L	0.33±0.15	1.14±0.16	2.24±0.81	0.21±0.11	0.97±0.06	0.14±0.04
	3000 mg/L	0.27±0.06	1.26±0.11	1.19±0.11	0.04±0.04	1.38±0.02	0.02±0.01
	5000 mg/L	0.30±0.13	1.22±0.11	1.34±0.19	0.06±0.00	1.19±0.12	0.02±0.01
EDTA	1000 mg/L	0.45±0.08	1.32±0.14	1.91±0.28	0.05±0.01	0.89±0.16	0.12±0.01
	3000 mg/L	0.42±0.14	1.16±0.21	1.85±0.04	0.04±0.00	1.90±0.00	0.13±0.04
	5000 mg/L	0.41±0.14	1.16±0.08	0.92±0.07	0.10±0.01	1.60±0.81	0.05±0.06
Alum	1000 mg/L	0.49±0.17	1.24±0.18	1.93±0.59	0.22±0.08	0.87±0.22	0.13±0.02
	3000 mg/L	0.55±0.11	1.01±0.04	1.06±0.55	0.19±0.08	0.26±0.09	0.16±0.02
	5000 mg/L	0.52±0.13	1.07±0.24	1.34±0.78	0.23±0.06	0.20±0.09	0.11±0.01
$FeCl_3$	1000 mg/L	0.35±0.01	0.64±0.71	1.91±0.02	0.18±0.01	0.90±0.04	0.14±0.01
	3000 mg/L	0.34±0.11	0.61±0.25	1.63±0.40	0.05±0.02	0.63±0.16	0.09±0.01
	5000 mg/L	0.24±0.13	0.90±0.12	0.95±0.12	0.17±0.02	0.51±0.57	0.09±0.08
$FeSO_4$	1000 mg/L	0.54±0.10	1.06±0.08	2.15±0.19	0.30±0.09	1.09±0.18	0.12±0.01
	3000 mg/L	0.54±0.15	1.39±0.08	2.19±0.54	0.30±0.09	0.78±0.21	0.20±0.02
	5000 mg/L	0.53±0.13	1.37±0.05	1.58±0.30	0.27±0.10	1.16±0.25	0.21±0.02
	NEMA [45]	0.1	1.0	10	0.1	1.0	1.0
	FEPA [41]	0.01	0.2	0.05	0.01	0.05	0.01

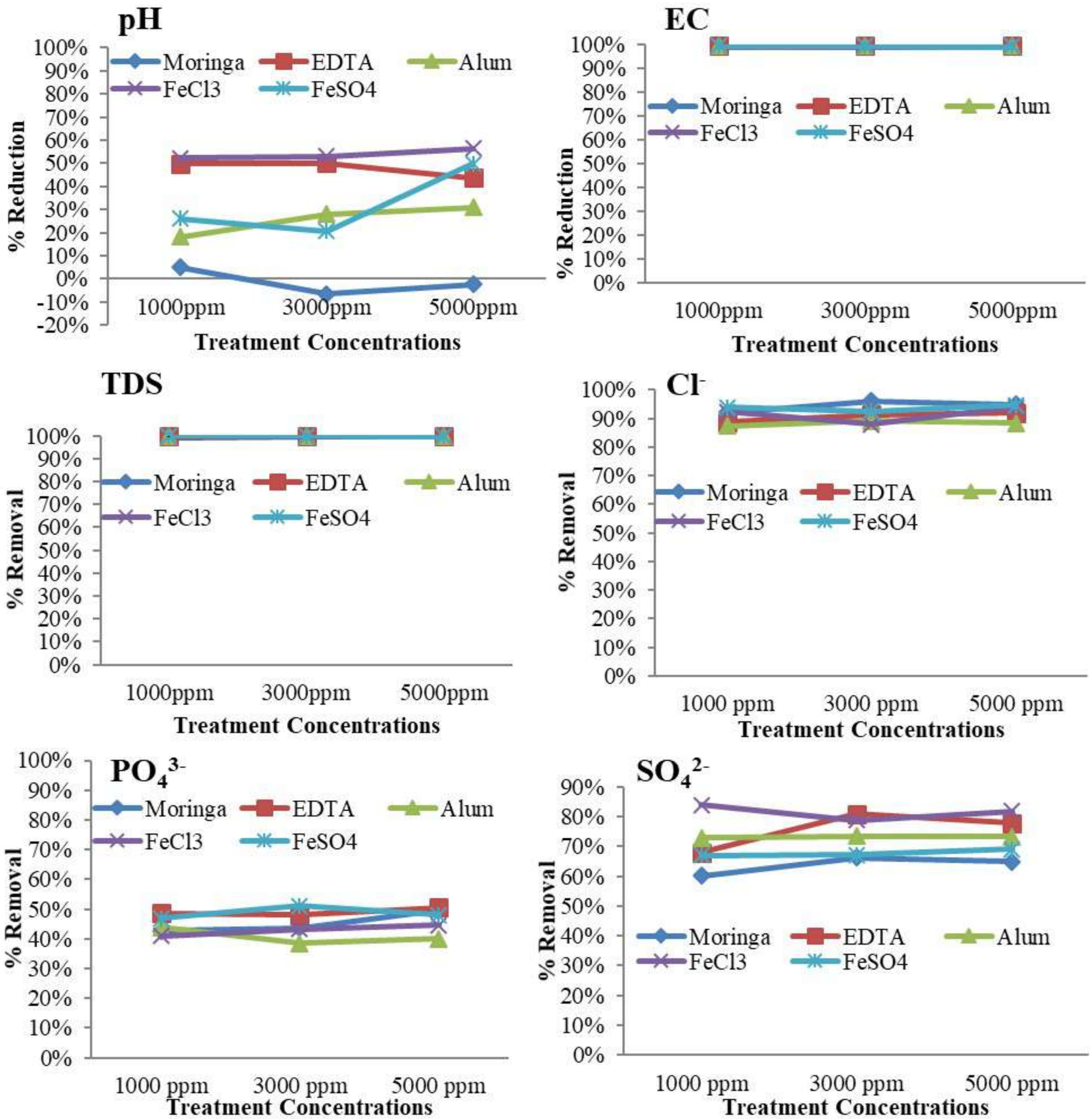

FIGURE 1

Removal/reduction of chemical parameters of leachates by coagulants and EDTA at varying concentrations

None of the coagulants and EDTA showed any preference for removal of EC and TDS. $FeCl_3$ showed highest affinity for pH reduction, while *Moringa oleifera* revealed the lowest reduction potential of pH. Even though, there was no much difference in the remediating abilities of these coagulants and EDTA in removing Cl^-; but it appeared that *Moringa oleifera* exhibited the highest removal efficiency. $FeSO_4$ and $FeCl_3$ also had slightly higher removing efficiencies for PO_4^{3-} and SO_4^{2-}, respectively. The effects of varying treatment concentrations of treatments were not obvious in the reduction of EC, TDS, Cl^- , PO_4^{3-} and SO_4^{2-}, except for $FeCl_3$ at 1000 mg/L, which showed highest percent removal of SO_4^{2-}.

Figure 2 presents the effects of coagulants and EDTA on metal concentrations of leachates. The coagulants and EDTA revealed varying remediating degrees at different doses of treatments. Furthermore, the removal affinity for particular metals has been clearly defined by the remediating agents. *Moringa oleifera* showed the highest removing ability for Ni and Cd, while $FeCl_3$ clearly reduced Cr in leachate samples than other coagulants. The percent removal of Cd by *Moringa oleifera* varied from 59-63%, similar to the study of Prasad and Rao [47] which documented removal efficiency of 70% for Cd in wastewater collected from Pradesh, India. Alum has the best removing potential for Mn as the remediating trend

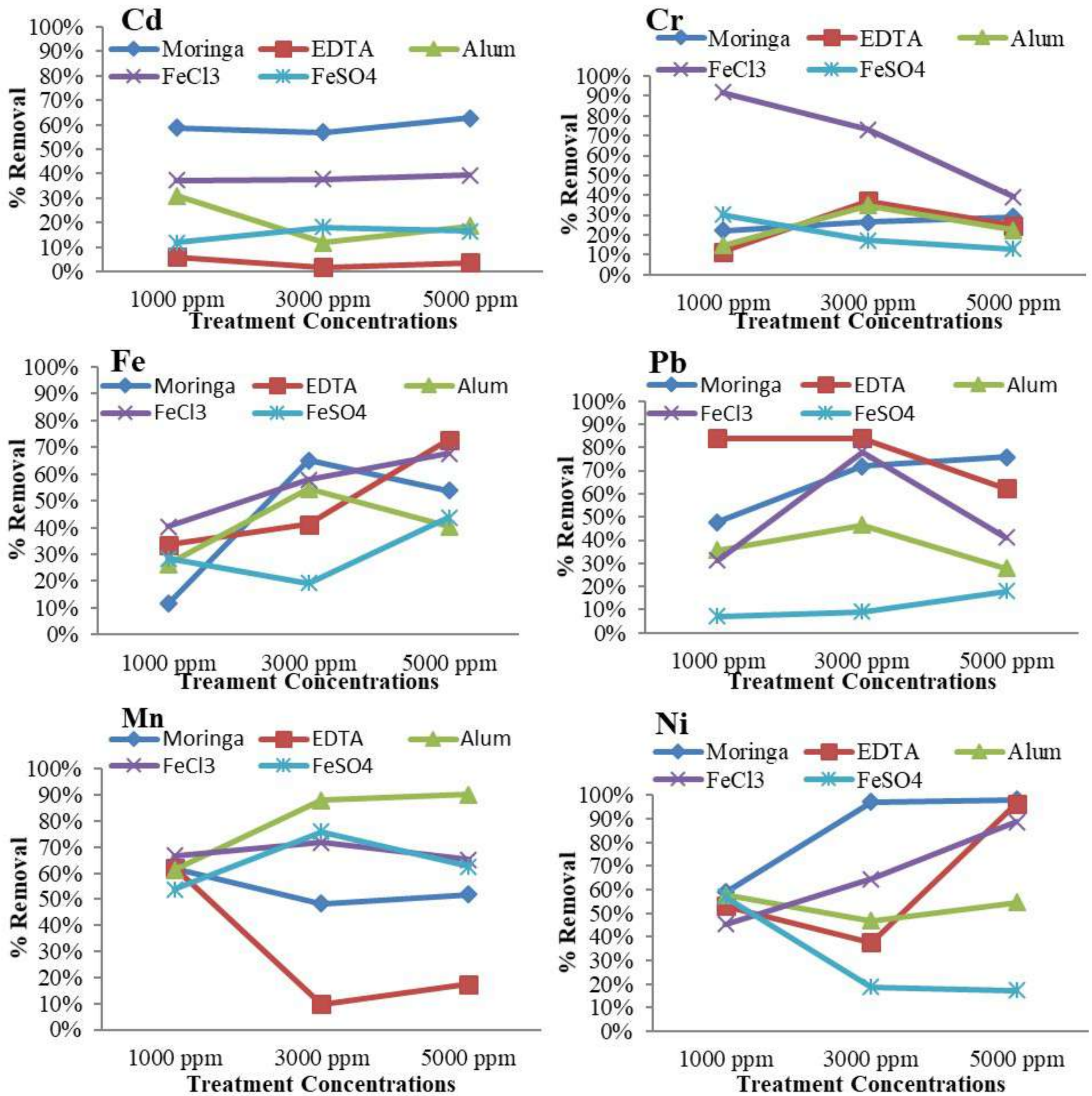

FIGURE 2

Metal removal by coagulants and EDTA at varying concentrations

increased with incremental treatment dosage. The EDTA was also very effective in removing Pb and Fe. Generally, the optimal removal of Cd, Cr and Fe by EDTA was at treatment 5000 mg/L. Similarly, the optimal reduction of Mn, Cd and Fe by $FeCl_3$ was observed at 5000 mg/L. Alum also removed the highest portion of Mn and Ni at 5000 mg/L. Previous studies have shown high percent removal of pollutants with increased dose of coagulants [48, 49].

Moringa oleifera was able to remediate 98% of Ni at 5000 mg/L, while EDTA also showed high removal efficiency of 97% for Ni at 5000 mg/L. $FeCl_3$ effectively removed 92% of Cr at 1000 mg/L, while Alum remediated up to 90% of Mn at 5000 mg/L. The study has revealed $FeSO_4$ as the least effective coagulant with removal efficiency varying between 7% for Pb at 1000 mg/L and 76% for Mn at 3000 mg/L. *Moringa oleifera* showed higher removing ability than many coagulants similar to the study of Eman *et al.,* [50] who demonstrated better removal of metals by *Moringa oleifera* than Alum. This study also showed more effective reduction of metals by $FeCl_3$ than Alum (except Mn), which is dissimilar to the studies of Ogunfowokan *et al.,* [51] and Yousefi and Zazouli [52].

The summary of metal removal potentials by different coagulants and EDTA is presented in Figure 3. The order of removal efficiency of metals followed $FeCl_3$ > *Moringa oleifera* > Alum >EDTA > $FeSO_4$. The optimum removal

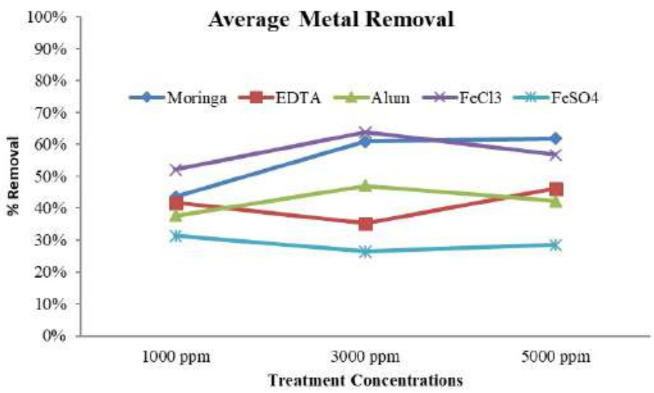

FIGURE 3
Summary of metal removal by coagulants and EDTA at varying concentrations

dose of metals by $FeCl_3$ and Alum was 3000 mg/L. *Moringa oleifera* and EDTA showed an optimum remedial capacity at 5000 mg/L, while for $FeSO_4$, the highest removal dose was 1000 mg/L. On the average, at treatment concentration of 1000 mg/L, metals reduced by 31% ($FeSO_4$) and 52% ($FeCl_3$), while at 3000 mg/L, removal efficiency of metals varied between 26% ($FeSO_4$) and 64% ($FeCl_3$). The coagulants and EDTA were able to remediate 28% ($FeSO_4$) to 62% (*Moringa oleifera*) of the mean total metals at 5000 mg/L.

CONCLUSION

The present study has remediated leachates collected from dumpsites in Abeokuta, Ogun state using four coagulants and EDTA. The remediating agents were prepared in the concentration range of 1000-5000 mg/L. The present study demonstrated the effectiveness of all the coagulants and EDTA in removing notable contaminants in dumpsite leachates. EC and TDS were completely removed from the leachates, while 87- 95% of Cl^- was remediated. The affinity of each remediating agent to reduction of metals was also observed. The EDTA has been effective for removal of Pb, $FeCl_3$ for Cr, Alum for Mn and *Moringa oleifera* for Ni and Cd. *Moringa oleifera*, EDTA and $FeCl_3$ were able to remediate Ni content in the leachates by 98, 97 and 89%, respectively. There were variations in the optimum removal doses of treatments. $FeCl_3$ and Alum were able to remove contaminants optimally at 3000 mg/L, *Moringa oleifera* and EDTA performed excellently at 5000 mg/L, while $FeSO_4$ was outstanding at 1000 mg/L. The sequence of removal efficiency of metals by remediating agents followed the decreasing order of $FeCl_3$ > *Moringa oleifera* > Alum >EDTA > $FeSO_4$. On the average, 31-64% of total metals in the leachates were removed by the four coagulants and EDTA indicating the promising potentials of coagulation and complexing methods.

ACKNOWLEDGEMENT

The authors are grateful to the Central Laboratory of University of Ibadan, where metal analyses were carried out. We are also grateful to the anonymous reviewers for their constructive criticisms.

Conflict of Interest

The authors declared none.

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