ADSORPTION STUDY OF DUMPSITE LEACHATE TREATMENT USING AGRICULTURAL WASTE AS BIOSORBENT

Comment [e1]: This topic is too vast.
The dumpsite should be known and the biosorbent (agricultural waste)

Comment [e2]: The authors are giving the impression of a bio sorption treatment which at the end, ends up with an activated carbon treatment.

ABSTRACT

Dumpsite leachate has the potential to pollute ground and surface water as well as vegetation within the vicinity of the dumpsite. Its treatment therefore needs adequate attention. The aim of this work is to study the adsorption of Lagos dumpsite leachate treatment using *Musa sapientum* peel as biosorbent with a view of establishing the adsorption isotherm model. *Musa sapientum* peels sourced from Ayetoro market in Epe area of Lagos State, Nigeria were used to prepare the adsorbent. Batch adsorption was carried out with various dosage of the prepared absorbent in leachate collected from Lagos dumpsite. The adsorption data obtained were fitted into Linear, Freundlich, Langmuir, Temkin and Hasley isotherm models. The results showed that the concentration of total dissolved solids (TDS) in the dumpsite leachate decreased as the adsorbent dosage increased. At adsorbent dosage of 10 g/L, the concentration of TDS in the leachate was 485.7 mg/L which was less than the 500 mg/L stipulated by National Environmental Standard and Regulatory Agency (NAESRA) for the discharge of wastewater. The coefficient of determination (R^2) values for Linear, Freundlich and Hasley, Langmuir and Temkin isotherm models were 0.9944, 0.9936, 0.8562 and 0.9723 respectively. Linear isotherm model was jettisoned because the plot did not pass through the origin and Freundlich isotherm model was ignored as a result of *N* value which was less than unity hence Hasley isotherm model was adopted in this work. A good correlation existed between the experimental and predicted values, having a R^2 value of 0.9965 which further validated the Hasley isotherm model as the best adsorption model for the treatment of Lagos dumpsite leachate using *Musa sapientum* peel as biosorbent. It was concluded that *Musa sapientum* peel as biosorbent can be used for treatment of Lagos dumpsite leachate.

Comment [e3]: Which isotherm are you talking of?

Comment [e4]: Consider revision
Or repetition.
Was activated carbon used or the musa sapientum peel as biosorbent??

Keywords: Biosorbent, adsorption, leachate, study and treatment.

Comment [e5]: I don't agree with the author that study should be consider as key word here

1. INTRODUCTION

When precipitation falls on wastes within the dumpsite or landfil site, it dossolves the soluble components of the wastes to form leachate. Leachate is a dark liquid which emits a strong odour (Norashiddin et al., 2019). It consists various pollutants such as heavy metals, organic substances, ammonia and inorganic salts (Uygur and Kargi, 2004; Renou *et al*., 2008; Aziz *et al*., 2009 and Foul *et al*., 2009). The leachate percolates through the soil matrix and pollutes the groundwater. It also pollutes surface water and affects vegetation. This makes it necessary for the leachate to be treated in order to avert the inherent dangers it poses.

Bioremediation is one way of treating leachate. It is a mean of destroying or rendering harmless variuos pollutants using natural biological activity. It involves the use of low – cost and low technological techniques (Akinyemi *et al*., 2019). Various types of agricultural wastes such as coffee, palm shell, bagasse, coconut shell, banana psedo – stem, rice husks and the host of others have been used in the treatment of landfill leachates (Ching *et al*., 2011; Lim, *et al*., 2009; Mohan and Singh, 2002 and Halm *et al*., 2010). It has been reported in the literature that the functional groups like hydroxyl, carbonxyl, thio and amino, phosphate present on the walls of agriculture wastes biomass play a very vital role for binding of contaminants which result in the treatment of landfill leachate (Deshmukh *et al*., 2017 and Kumar *et al*., 2011).

Comment [e6]: Articles should be sited either according to year or in alphabetical order.

Comment [e7]: Agricultural

Extensive works have been done on the treatment of leachates by scholars (Foo *et al*., 2013; Lim *et al*., 2010; Aziz *et al*.,2011; Kalderis *et al*., 2008; Liyan *et al*., 2009; Ab Ghani *et al*., 2017; Zajc *et al*., 2004; Hur and Kim, 2000; Dabrowski, 2001; Aghamohammadi *et al*., 2007 and Yahya *et al*., 2017). Kaideris *et al*. (2008) worked on adsorption of polluting substances on activated carbon prepared from rice husk and suger cane bagasse. The work revealed 70 and 60

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percent removal of chemical oxygen demand (COD) and colour respectively. Aziz *et al*. (2011) carried out landfill leachate treatment using powdered activated carbon augmented sequencing batch reactor process. It was shown that 64.1, 71.2 and 81.4 percent removal of COD, colour and ammoniacal nitrogen were achieved. Foo *et al*. (2013) used banana frond activated carbon prepared by microwave induced activation to remove boron from landfill leachate. 92.73 percent of the boron were removed from the leachate. Ab Ghani *et al*. (2017) investigated the optimization of preparation conditions for activated carbon from banana pserudo – stem using response surface methodology on the removal of colour and COD from landfill leachate. The investigation showed that 91.2 and 83 percent removal of colour and COD respectively.

Rodrigo *et al*. (2019) research on landfill leachate treatment using activated carbon obtained from coffee waste. The research revealed that the ammonia adsorption data obtained were successfully modelled by Freundlich isotherm. It is evident from the myraid of literature that works on removal of total dissolved solids (TDS) especially from Lagos dumpsite leachates are very scarce. Therefore the aim of this work is to study the tretment of Lagos dumpsite leachate using *Musa sapientum* peels as biosorbent with a view of establishing the adsorption isotherm model which provides useful information on the adsorption capacity of the biosorbent. The adsorption model is an important tool for predictiong the design of adsorption batch reactor. The treatment of Lagos dumpsite leachate will minimise the inherent dangers pose by the leachate which justifies the work.

Muse sapientum peels were chosen for the treatment of Lagos dumpsite leachate because of the presence of free hydroxyl group of polymeric compounds such as lignin or pectin that contain the functional groups of alcohols, phenols and carboxylic acids and the N – H bending vibration of primary amines in the walls of the *Muse sapientum* peels agricultural waste as established in the work of DesMukh *et al*. (2017). The scanning elctron microscope of *Muse sapientum* peel

Comment [e9]: Do we have only one dumpsite in Lagos? If no then the author need to be precise

Comment [e10]: ??

surface adopted from the work of Alaa El – Din et al. (2018) and the Fourier transformation infrared spectroscopy (FTIR) obtained from the work of DesMukh *et al*. (2017) are shown in Figures 1 and 2 respectively. *Muse sapientum* peels were also used in this work as a result of their low cost and lack of alternative use.

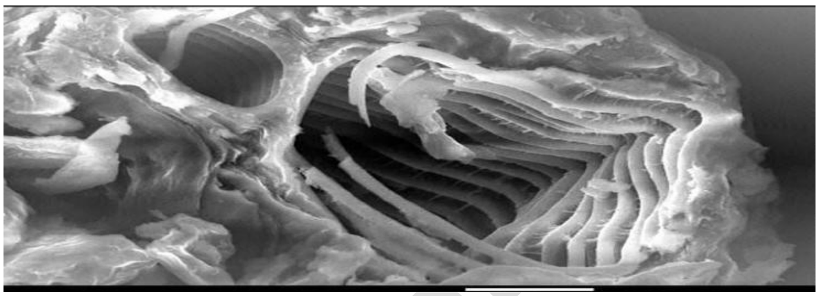

Fig. 1. Scanning electron microscopeof Muas sapientum peel surfae

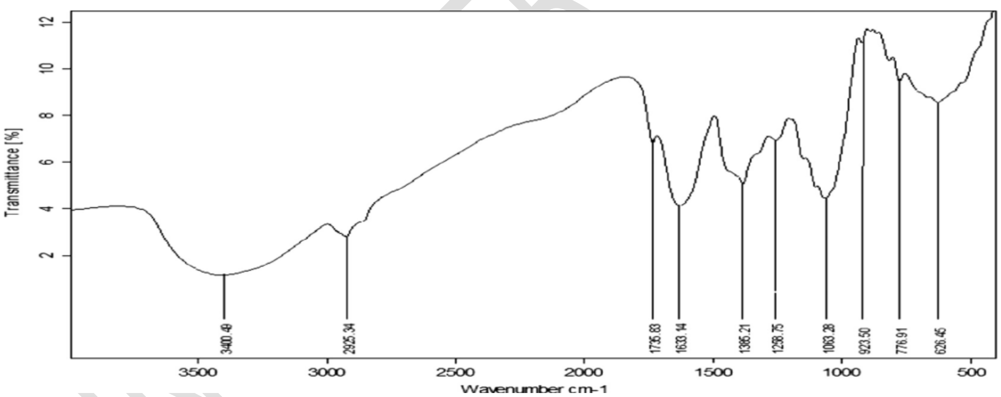

Fig. 2. FTIR spectra of *Musa sapientum* peel

2. METHODOLOGY

2.1 Preparation of Adsorbent

Muse sapientum peels were sourced from Ayetoro market in Epe area of Lagos State, Nigeria. The peels were cleaned with water to remove any undesrable materials. The peels sample were carbonised in a furnace for a duration of 1 hr. The char product from the furnace was quencehed

Comment [e11]: I don't see the need of figure 1 and 2 in this section of work.

Comment [e12]:

Comment [e13]: Carbonization mean changing from organic nature to carbon at high or moderate temperature.
What was the carbonization temperature?

Comment [e14]: Equilibrium or resting time is an important factor. I don't know why the author decided to fixed it at 1h?

with cold water in order to cool and then tranferred into the oven for further drying at 110^oC. The activated carbon was impregnated with tetraoxosulphate (vi) (H_2SO_4) (10 % by weight) followed by heating in the absence of air. The resultant moist paste was charged into the furnace and heated for 1 hr at a temperature of 110^oC until a constant weight of activated carbon was achieved. The chemical activation was carried out to remove the tar from the pores of the activated carbon. The activated carbon was rinsed thoroughly with distilled water to remove the remaining H_2SO_4. The activated carbon was then dried in an oven at a temperature of 110^oC for 3 hrs. The refined activated carbon was crushed with a mortar to size of 100 mesh.

2.2 Batch Adsorption

The method of Olafadehan *et al*. (2012) was adopted with a little modification. Batch adsorption equilibrum studies were carried out using the produced activated carbon of different known masses. Each mass was placed in 250 ml Erlenmeyer flask each containing 100 ml of leachate collected from Soluos dumpsite in Lagos State, Nigeria having a pH of 7.2. The flasks were agitated continuously at agitation speed of 150 rpm for 2 hrs by which equilibrum must have occured. The resultant mixture was filtered and the concentrations of TDS in the filtrates were determined as compared to COD in the work of Olafadehan *et al*. (2012). The concentrations of the TDS were determined according to the standard methods for examination of water and waste water as prescribed by American Public Health Association (APHA, 1994). The adsorption capacity values at equilibrum, q_e [=] mg/g, were evaluated using Equation (1).

$$q_e = \left(\frac{c_o - c_e}{m}\right)V = \frac{X}{M} \qquad (1)$$

Where c_o is the initial adsorbate concentration [=] mg/L and c_e is the adsorbate concentration at equilibrium [=] mg/L, m is the mass of adsorbent [=] g, V is the volume of aqueous solution (the

Comment [e15]: The cooling system here is not the best. Scientifically cooling is usually done in a desiccator. Why using cold water, and if cold as presume, what is the temperature? And was it mixed with the carbon/char or it was kept in a container and inserted into the water?

Comment [e16]: You can't impregnate the activated carbon again.
You can only activate the chars to activated carbon
So what are you talking of?

Comment [e17]: The choice of 10% by weight of sulphuric acid must be known

Comment [e18]: How can you justify this?

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Comment [e19]: ?

Comment [e20]: ???

Comment [e21]: English problem

Comment [e22]: The author can't go ahead for adsorption without characterizing the activated carbon.
The surface area which is one of the most important factor in adsorption must be presented.
Pore volume and size also
The surface phenomenon, which deals with functional groups must also be known

Comment [e23]: We expect the complete analyzed sample of leachate before adsorption and after absorption. Not only the pH

Comment [e24]: If it is a comparison work with what was done in 2012, I don't think it is right for the dumpsite activity is not constant

Never can the TDS be constant throughout the years

Which means that the author must look for ways to do the analysis as required scientifically leachate in contact with the adsorbent) [=] L, X the change in initial concentration and equilibrium concentration [=] mg/L and M is the carbon dosage [=] g/L.

N

Comment [e25]: How was the residual concentration determined? By using which method and how?

3. RESULTS AND DISCUSSION

The analysis of batch adsorption equilibrium data for Lagos dumpsite leachate sample using *Musa sapientum* peel activated carbon at temperature of 30^{o}C is presented in Table 1. The concentration of TDS in the dumpsite leachate decreased as the adsorbent dosage increased. At adsorbent dosage of 10 g/L, the concentration of TDS in the leachate was 485.7 mg/L which was less than the 500 mg/L stipulated by NAESRA for the discharge of wastewater.

Comment [e26]: Note here that carbon can not be produce at 30 degree.

Table 1. Analysis of batch adsorption equilibrium data for Lagos dumpsite leachate sample using *Musa sapientum* peel activated carbon at temperature of 30^{o}C

Activated carbon dosage (g/100 ml)	Activated carbon dosage, M (g/L)	TDS, c_e (mg/L)	X (mg/L)
0	0	2101.7	-
0.2	2	1100	1001.7
0.4	4	811	1290.7
0.6	6	632	1469.7
0.8	8	529	1572.7
1.0	10	485.7	1616

Comment [e27]: Why using the two unit?

Table 1. Analysis of batch adsorption equilibrium data for Lagos dumpsite leachate sample using *Musa sapientum* peel activated carbon at temperature of 30^{o}C. (Continuation)

q_e = (X/M), (mg/g)	*ln* c_e	*ln* q_e	C_e/q_e (g/L)
-	-	-	-
500.85	7.003	6.2163	2.196
322.675	6.698	5.777	2.513
244.95	6.449	5.5011	2.580
196.588	6.271	5.281	2.691
161.6	6.186	5.085	3.006

Sorption equilibrium data generated in the treatment of Lagos dumpsite leachate were tested with different adsorption isotherm model given in Equations 2, 3, 5, 12 and 13.

- 1 – p Henry law: The one – parameter (1 - p) Henry law also known as linear adsorption isotherm is used to describe adsorptions at relative low pressure (Olafadehan, 2021). It is of the form shown in Equation (2) for liquid – phase adsorption.

$$q_e = k'_H c_e \qquad (2)$$

Where k'_H is the Henry law constant which was determined by plotting q_e against c_e. If a straight line passing through the origin was obtained with regression coefficient (R^2) very close to unity, it means the isotherm model can be used to correlate the experimental data.

- 2 – p Freundlich isotherm: The two – parameter (2 - p) Freundlich isotherm describes adsorption on heterogeneous surfaces or surfaces supporting sites of varied affinities (Olafadehan, 2021). It also illustrates a multilayer adsorption which assumes the stronger binding sites are occupied first and the binding strength decreases with increasing degree of adsorption (Ewecharoen *et al*., 2008). The Freundlich isotherm is presented in Equation (3).

$$q_e = k_F c_e^{\frac{1}{N}} \quad (3)$$

Where k_F is a constant which indicates the Freundlich adsorption capacity and parameter, *N*, characterizes the homogeneity of the system. Taking the logarithm of Equation (3) yields:

$$\ln q_e = \ln k_F + \frac{1}{N} \ln c_e \quad (4)$$

The Freundlich rate coefficient, *N* and k_F were evaluated from the slope and intercept on q_e axis respectively when $ln\ q_e$ was plotted against $ln\ c_e$. If a straight line is obtained having a very high R^2 close to unity. Freundlich isotherm can be used to correlate the adsorption data of a system provided $N \succ 1$. The adsorption is considered favourable provided $0 \prec \frac{1}{N} \prec 1$ which indicates chemisorption and taken as unfavourable when $\frac{1}{N} \succ 1$.

- 2 – p Langmuir isotherm: The 2 – p Langmuir isotherm describes the monolayer adsorption on a solid surface where all sites are equivalent and independent. For liquid phase adsorption, the model is given as shown in Equation (5) (Olafadehan, 2021).

$$q_e = \frac{q_{max} K_L c_e}{1 + K_L c_e} \quad (5)$$

Where q_{max} is the Langmuir constant related to the adsorption capacity (maximum adsorption capacity for solid phase loading) [=] mg/g and K_L is the energy constant related to the heat of adsorption [=] dm^3/mg. The various linear forms of Equation (5) are presented in Equations (6) – (10).

Comment [e28]: The author is only using Olafadehan as reference which makes research very difficult

$$\frac{1}{q_e} = \frac{1}{K_L\, q_{\max}} \frac{1}{c_e} + \frac{1}{q_{\max}} \tag{6}$$

$$\frac{c_e}{q_e} = \frac{1}{q_{\max}} c_e + \frac{1}{K_L\, q_{\max}} \tag{7}$$

$$q_e = \frac{1}{K_L} \frac{q_e}{c_e} + q_{\max} \tag{8}$$

$$\frac{q_e}{c_e} = -K_L q_e + K_L q_{\max} \tag{9}$$

$$\frac{1}{c_e} = -K_L q_{\max} \frac{1}{q_e} - K_L \tag{10}$$

The plot of Langmuir isotherm model was generated with the use of Equation (7) since it has been established in the work of Olafadehan *et al*. (2018) that Equations (6) – (10) gave almost the results. Hall *et al*. (1966) and Malik (2004) descibed the important characteristics of Langmuir isotherm using a dimensionless constant, R_L, also known as separation factor or equivalent parameter and is given by Equation (11).

$$R_L = \frac{1}{1 + K_L\, c_o} \tag{11}$$

Where c_o is the initial concentration of adsorbate. Zhai *et al*. (2004) showed that R_L is an indication of the shape of the isotherm thus $R_L \prec 0$, $0 \prec R_L \prec 1$, $R_L = 1$ and $R_L \succ 1$ represent irreversible, favourable, linear and unfavourable isotherm respectively.

- 2 – p Temkin isotherm: The 2 – p Temkin isotherm reveals the effects of indirect adsorbent – adsorbate interactions on the adsorption process. For liquid phase adsorption, the Timkin isotherm model is shown in Equation (12).

$$q_e = \frac{RT}{b_T}\ln\left(A_T\, c_e\right) = \frac{RT}{b_T}\left(\ln A_T + \ln c_e\right) \qquad (12)$$

Where b_T is the Temkin constant, which is related to the heat of adsorption [=] J/mol, A_T is the Temkin isotherm constant [=] L/g. A plot of q_e against lnc_e gives a straight line with slope equals $\frac{RT}{b_T}$ and intercept on q_e axis equals $\frac{RT}{b_T}\ln A_T$. These make the b_T and A_T to be determined at the adsorption isotherm.

- Hasley isotherm: This is important in the evaluation of multilayer adsorption at a relatively large distance from the surface (Ayawei, 2015). The isotherm is presented in Equation (13) and when linearised yields Equation (14).

$$q_e = \left(K_H c_e\right)^{\frac{1}{n_H}} \qquad (13)$$

$$\ln q_e = \frac{1}{n_H}\ln K_H + \frac{1}{n_H}\ln c_e \qquad (14)$$

Where K_H and n_H are the Hasley isotherm constants. A plot of $ln\ q_e$ against $ln\ c_e$ was made. This would yield a straight line graph with slope equivalent to $\frac{1}{n_H}$ hence n_H and K_H can be determined.

Table 2 shows isotherm parameters for the batch adsorption equilibrium data while Table 3 reveals the experimental (actual) and predicted values as well as percentage difference for Freundlich and Hasley isotherm models.

Table 2.Isotherm parameters for batch adsorption equilibrium data

Isotherm	Parameters	Value

Linear	R^2	0.9944
	k_H	0.536
Freundlich	R^2	0.9936
	N	0.7551
	k_F	0.0467
Langmuir	R^2	0.8562
	q_{max}	-909
	K_L	0.0003
	R_L	0.593
Timkin	R^2	0.9723
	b_T	6.253
	A_T	519.569
Hasley	R^2	0.9936
	n_H	0.7551
	K_H	0.0989

Table 3. Actual and predicted values as well as percentage differences for Freundlich and Hasley isotherm models

M (g/L)	q_e (mg/g)	Freundlich model		Hasley model	
		q_e (Predicted) (mg/g)	% difference	q_e (Predicted) (mg/g)	% difference
2	500.85	497.775	0.614	497.80	0.609
4	322.675	332.455	- 3.031	332.472	- 3.036
6	244.95	238.95	0.0124	2.445	2.445

8	196.588	188.795	0.04	3.96	3.96
1.0	161.61	168.606	- 0.043	- 4.34	- 4.34

Figures 3 – 6 depict the Linear, Freundlich and Hasley, Langmuir and Temkin isotherm models respectively for the treatment of Lagos dumpsite leachate while Figure 7 reveals the correlation between the predicted and actual values for Hasley isotherm model.

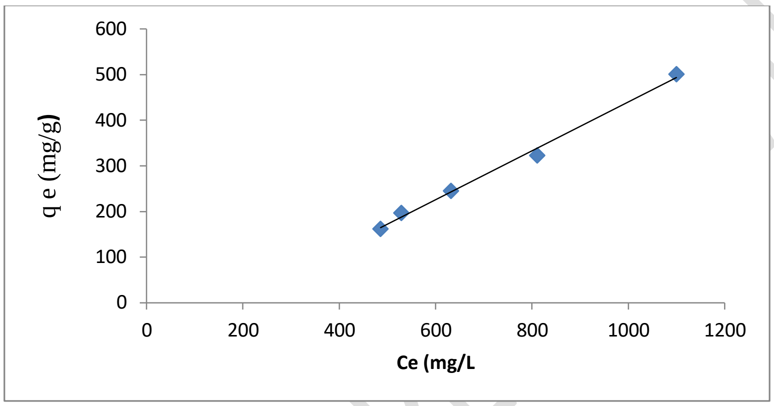

Fig.3. Linear isotherm plot for treatment of Lagos dumpsite leachate.

Comment [e29]: This straight line curve must not start at zero point. For good presentation as shown, it should start at Ce = 400 as the initial point to avoid empty spacing

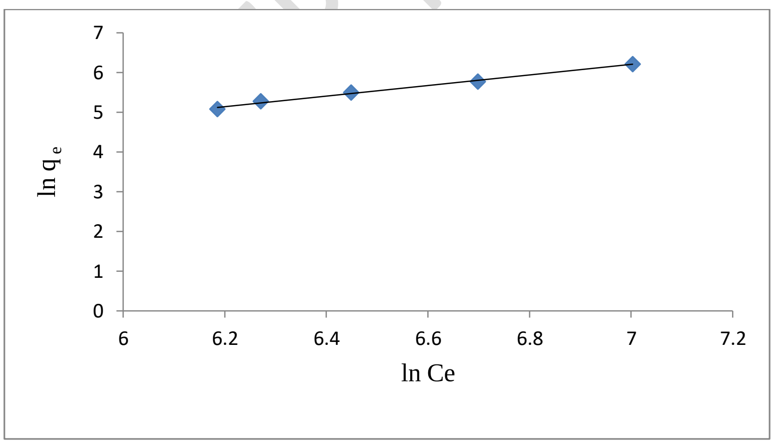

Fig.4. Freundlich and Hasley isotherms plot for treatment of Lagos dumpsite leachate.

Comment [e30]: Same here for the y-axis, it should cut at 4

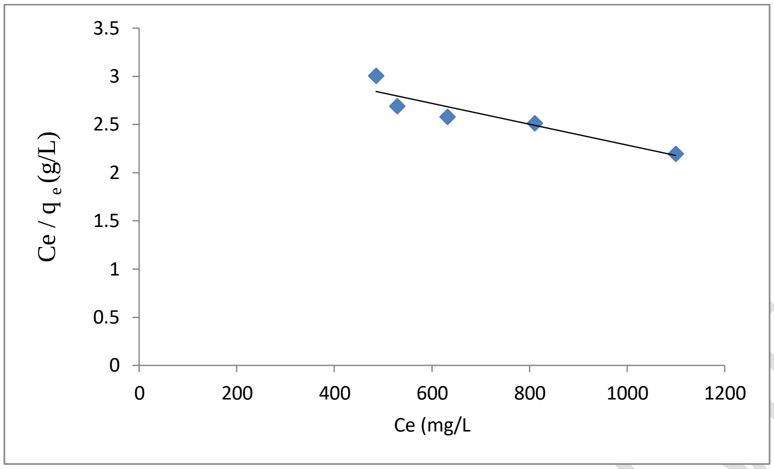

Fig.5. Langmuir isotherm plot for treatment of Lagos dumpsite leachate.

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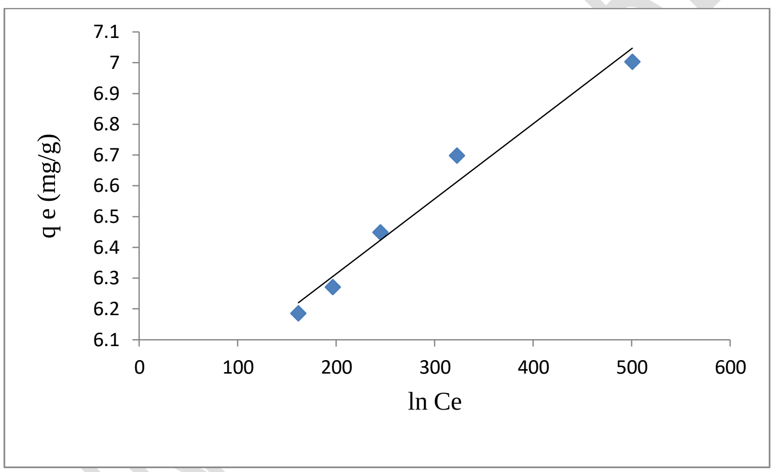

Fig.6. Timkin isotherm plot for treatment of Lagos dumpsite leachate.

Comment [e32]: Qe = 100 + for the minimum point

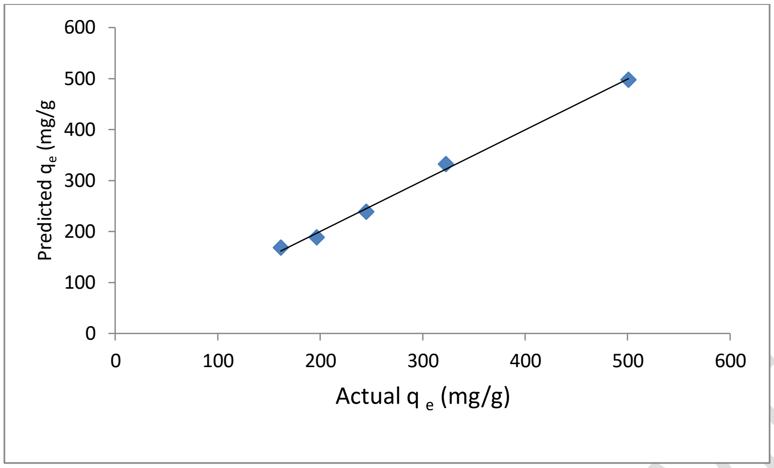

Fig.7. A graph of predicted against actual values for Hasley isotherm model

Comment [e33]: Qe = 100 +

The R^2 value for linear, Freundlich and Hasley, Langmuir and Temkin isotherm models were 0.9944, 0.9936, 0.8562 and 0.9723 respectively. The closer the R^2 value to unity, the better the fitness of the experimental data to the isotherm model. Linear isotherm model has the highest R^2 value followed by Freundlich and Hasley isotherm models which have the same R^2 value 0.9936. For the linear isotherm model to be chosen based on its R^2 value, the plot of q_e against c_e must pass through the origin which in this case, did not hence the linear isotherm model was jettison. The isotherm parameters of Freundlich and Hasley were substituted into the models which were then used to predict the experimental data. The percentage difference in absolute term between the experimental and predicted data for Freundlich and Hasley isotherm models ranged between 0.0124 and 3.031 and between 0.609 and 4.34 respectively which indicated Freundlich isotherm model was better in term of percentage difference between the experimental and the predicted data. However, for the Freundlich isotherm model to represent adsorption data, the value of N must be greater than unity and in this case, the value of N was 0.7551 which was less than unity hence Freundlich isotherm model was ignored and Hasley isotherm model was adopted as the isotherm model for the treatment of Lagos state dumpsite leachate using *Musa sapientum* peel as biosorbent. A good correlation existed between the predicted and actual values as shown in Figure 7 for Hasley isotherm model, having a R^2 value of 0.9965. This further validated that the Hasley isotherm model correlated the experiment data obtained for the treatment of Lagos dumpsite leachate using *Musa sapientum* peel as biosorbent. Hasley isotherm model has also been applied to the removal of lead (11) from aqueous solution using shell carbon prepared by KOH (Song *et al*., 2013). The Hasley model fitted the experimental data with a high R^2 value, which was attributed to the heterogeneous distribution of active sites and multilayer adsorption of the shell carbon (Ayawei, 2015).

4. CONCLUSION

The equilibrium adsorption study of treatment of Lagos dumpsite leachate with a view of establishing the adsorption isotherm model which provides useful information on the adsorption capacity of the adsorbent has been carried out. The concentration of TDS in the dumpsite leachate decreased as the adsorbent dosage increased. At adsorbent dosage of 10 g/L, the concentration of TDS in the leachate was 485.7 mg/L which was less than the 500 mg/L stipulated by NAESRA for the discharge of wastewater. Linear isotherm model has R^2 value of 0.9944 which was greater than the R^2 value of Hasley isotherm model but it was jettison because the plot did not pass through the origin. Hasley and Freundlich isotherm models have the same R^2 values but Freundlich isotherm model was ignored as a result of N value which was less than unity hence Hasley isotherm model was adopted to have fitted the adsorption data exceedingly well. A good correlation existed between the experimental and the predicted values, having a R^2 value of 0.9965 which further validated the Hasley isotherm model as the best adsorption model for the treatment of Lagos dumpsite leachate using *Musa sapientum* peel as biosorbent. Hence *Musa sapientum* as biosorbent can be used for treatment of Lagos dumpsite leachate.

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Comment [e39]: The authors are very negligence

Spellings should be verified

English should be reexamine