IOSR Journal of Environmental Science, Toxicology and Food Technology (IOSR-JESTFT)
e-ISSN: 2319-2402,p- ISSN: 2319-2399.Volume 12, Issue 4 Ver. I (April. 2018), PP 26-33
www.iosrjournals.org

Adsorption and Equilibrium Studies of Textile Effluent Treatment with Activated Snail Shell Carbon

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Abstract: *This paper investigated the ability of snail shell-derived activated carbon adsorbent to adsorb Color, Total Suspended Solids (TSS) and Mg hardness from textile industry wastewater. The effect of adsorption treatment on some physico-chemical parameters viz: pH, Total hardness, Conductivity, Salinity and Total Dissolved Solids (TDS) was examined as well. Batch adsorption experiment in Jar Apparatus was carried out by varying adsorbent dosage from 1g – 6g (1g increments) in 200ml of wastewater sample and stirring at constant agitation speed of 120 rpm for 1hr contact time at room temperature for better mass transfer. The results showed maximum removal efficiencies of 88.9% and 87.1% at 30g/l dosage for color and TSS respectively and 72% at 20g/l for Total hardness. The pH of treated effluent reduced slightly to 8.0 from 8.4 at maximum dosage of 30g/l. However, TDS, Conductivity and Salinity increased correspondingly with dosage. Ca hardness increased with dosage amount up to 20g/l before reducing slightly. High coefficient of correlation, R^2 ranging between 0.85 and 0.94 were obtained for colour and TSS removal by Freundlich and Langmuir linear isotherm models. This proves the applicability of both isotherm models to describe experimental process.*

Keywords: *Activated carbon, Adsorption, Textile Wastewater, Snail Shell, Isotherm models.*

Date of Submission: 05-03-2018 Date of acceptance: 26-03-2018

I. Introduction

The textile industry has been criticized as one of the world's worst offenders in terms of pollution because of heavy usage of three components viz: chemicals, dyes and water [2]. Large amount of wastewater is generated from the various processing units such as: desizing, scouring, bleaching, mercerizing, printing and dyeing. The main problems with textile industry wastewater are high Biological Oxygen Demand (BOD), Chemical Oxygen Demand (COD), strong color residual, highly fluctuating pH and other solution substances. When compared to other parameters, the removal of BOD, COD and color pose the greatest environmental concern. Color removal from textile effluents has attracted attention not only because of its potential toxicity, but mainly due to its visibility problems [21].

Numerous effluent treatment methods, such as advanced oxidation processes [19], ozonation [11], electro-oxidation [8], photochemical oxidation using UV/H_2O_2 [30], electrochemical techniques [9], coagulation–flocculation [26], ion exchange [10] and membrane processes [7] among others have been used to treat various industrial effluents. However, many of these technologies are cost-prohibitive, especially when adopted by small-scale textile industries to treat large streams of wastewater. Among these methods, adsorption technique seems to have the most potential for future use in industrial wastewater treatment due to its proven efficiency in the removal of organic and mineral pollutants and for economic considerations [1, 6].

Adsorption is one of the established unit operations used for the treatment of contaminated water [24] and are usually conducted over batch and column studies. Activated carbon is the most used adsorbent. Due to its high cost and considering the enormous quantity of effluent produced by textile industries, researches are turning toward the use of alternative adsorbents also called "non-conventional low-cost adsorbents" [12]. The cost of producing activated carbon from inexpensive and readily available materials is often less compared to the commercial activated carbon and offers promissory environmentally-friendly benefits.

Some of the activated carbons used to treat industrial wastewater in the recent past are corncob, groundnut husk, rice husk, tea leaves carbon, saw dust [20] eucalyptus bark [5] cattle horn and bone [22] spent activated carbon [29] among others. Investigators have also examined a wide variety of adsorbents like fly-ash [23], peat, sawdust, brown coal and bagasse to remove colour from textile industry wastewater. In line with the quest to discover cheap and readily available adsorbent, this study attempts to investigate the capability of Africa Giant snail (*Archatina archatina*) shell which is grossly under-utilized in Nigeria as an adsorbent for removal of color and detoxification of textile wastewater.

II. Material and Methods

2.1 Study Area

Abeokuta, the capital of Ogun State, Nigeria is the project area under investigation. Abeokuta is a historic town founded by the Egbas in 1830 and situated in the southwestern part of the Federal Republic of Nigeria. It is 100km north of Lagos and 80km southwest of Ibadan. It is located in the sub-humid tropical region of Southwestern Nigeria (Latitudes 7°5' N to 7°20' N and Longitudes 3°17' E to 3°27'). The mean annual rainfall and temperature are about 1,270 mm and 28°C respectively while the estimated mean annual potential evaporation is 1,100 mm.

The major site of local textile production is Itoku situated in Abeokuta south local Government area of Ogun state. Itoku is renowned for the production of "adire" and "kampala" textile types. However, production is majorly manual with no stringent environmental regulatory standards in place to control the activity of the local textile industries. Consequently, the indiscriminate discharge of untreated effluents by these dyeing units and local industries have impacted severely on quality of nearby waterbodies, groundwater and shallow wells. Hence, this research is aimed at production of low-cost point-of-use (POU) adsorbent from readily available snail shell waste for pretreatment of these local textile industries effluent before discharge.

2.2 Preparation of Activated Snail Shell Adsorbent

The Snail Shells that was used for this study were collected from Bodija market in Ibadan, Oyo State, Nigeria. The procedure by Grigis and Ishak, [13] with slight modifications was used. The shells were washed properly in and out to remove dirt. The washed shells were dried first in open air and intense sunlight for about 2hrs and then put in an oven for another 2hrs at a temperature of 110^{o}C. The precursor was wrapped in double layer of aluminum foil to facilitate deoxygenated condition and carbonized in electric muffle furnace at carbonation temperature of 350^{o}C for 3 hours. The carbonized shells were then crushed and sieved to obtain sizes smaller than 600µm. Thereafter, 300 g of the carbonized sample was mixed with 20% of the solvent (H_3PO_4) solution at an impregnation ratio of 1:1 by weight. The impregnation was done at 80°C by heating the acid impregnated snail shell precursor mixture on electric stove and continuous stirring until slurry was formed. The slurry was then subjected to a temperature of 500^{o}C for 3 hours in electric muffle furnace for proper activation and cooled to room temperature. The activated sample was then washed repeatedly with deionized water to ensure there was no trace of excess acid in the activated sample. It was dried overnight at 120°C in oven and stored in air-tight jar for further studies.

2.3 Collection of wastewater

Grab sampling method was the mode for collecting the wastewater for this study. The wastewater was collected in 5L jerry can and capped plastic containers which were properly washed using distilled water and allowed to dry. The containers were rinsed three times with wastewater before filling to the brim with wastewater samples and firmly capped. The samples were transported under iced condition to inhibit ageing effect and changes in physico-chemical properties of wastewater. To avoid biodegradation, the sample was preserved in a refrigerator at 4^{o}C.

2.4 Analytical Procedures

Color which was reported in Hazen Units (HU) was determined using Nesselerizer Lovi-bond cuvette. The disk was rotated for color matching and the values were recorded. pH was determined using a glass electrode pH meter while Universal testing meter (Exstik II) was used to determine TDS, Salinity and Electrical Conductivity. The other physico-chemical parameters of TSS and Total hardness were determined according to Standard Methods for the Examination of Water and Wastewater [4]. All the adsorption experiments were performed in triplicate to ensure representative and reproducible results.

Experimental

2.5.1. Adsorption Experiments

Batch adsorption experiments were performed by contacting varying adsorbent dosages (1g to 6g) in increment of 1g with 200ml of textile wastewater sample at natural wastewater pH of 8.4. The experiments were performed in Jar Test Apparatus for a period of 1 hr at 120 rpm using 500 ml Erlenmeyer flasks containing wastewater samples at room temperature (30^{o}C). Continuous mixing was provided during the experimental period with a constant agitation speed for better mass transfer with high interfacial area of contact. The residual contaminants concentrations in each sample after adsorption at different adsorbent dosage was determined after filtering the treated wastewater sample with Whatmann filter paper No. 44 to make it adsorbent-free.
The capacity of adsorbent (q_e) was calculated using the following equation [31]:

$$q_e = \left(\frac{C_o - C_e}{W}\right) V \quad (1)$$

The reduction efficiency (RE) was calculated by the following equation [16]:

$$RE\ (\%) = \left(\frac{C_o - C_e}{C_o}\right) * 100 \quad (2)$$

where, c_0 and c_e are the initial and after treatment concentrations, respectively; W is the weight of the adsorbent used (g) and V is the volume of wastewater sample (l).

III. Results and Discussion

3.1 Adsorbent dosage effect on removal of contaminants

Table 1 below shows the result of characterization of raw textile effluent before adsorption treatment with prepared snail shell derived activated carbon.

Table 1: Raw textile wastewater physico-chemical characteristics All units in mg/l unless otherwise stated

Parameter	Average Value
pH	8.4
Color, haze	450
Total Suspended Solids	3200
Total Dissolved Solids	1750
Electrical Conductivity, µS/cm	2130
Salinity	1110
Total hardness, $CaCO_3$	2500
Ca hardness	125
Mg hardness	2375

Figure 1 shows the effect of different dosages of activated snail shell adsorbents on percentage removal of colour, TSS, Total hardness and Mg hardness. The percentage removal with increasing adsorbent dosage (5g/l – 30g/l) increased from 51.1 – 88.9%, 46 – 87.1 %, 65.5 – 72% and 76.3 – 95.8% for color, TSS, Total hardness and Mg hardness respectively. Figure 2 shows the effect of varying adsorbent dosage on pH of treated textile wastewater.

The uptake of color, TSS and Total hardness increased rapidly with increase in adsorbent dosage. This can be attributed to increased adsorbent surface area and availability of more adsorption sites. This is in line with findings of [15] on removal of color from textile effluent and [27] findings where an increasing amount of activated snail shell adsorbent dosage resulted in corresponding diminution of TS and TSS in beverage effluent.

The removal of these contaminants can also be attributed to favourable pore distribution and electrically unsaturated sites achieved through chemical activation with H_3PO_4. This increased the capacity of the adsorbent to remove both colloidal and suspended particles, colouring agents, carbonates and hydroxides which are present in the raw textile effluent. The rate of removal of pollutants at 5 – 15g/l dosage of adsorbent is much higher than the 15 – 30g/l range. This may be due to overlapping of adsorption sites as a result of overcrowding of adsorbent particles. This agrees with the finding of [15, 17].

The pH of the treated effluent reduced with corresponding increase in adsorbent dosage. This is due to the removal of weak and strong base such as carbonates, bicarbonates and hydroxides which were initially present in the raw effluent by the activated snail shell adsorbent. Also, the presence of residual acid (H_3PO_4) used for activation may be responsible for the slight reduction in pH of treated effluent. This is line with the finding of [27].

The result showed substantial removal of Total hardness with increasing adsorbent dosage from 5g/l to 20g/l after which increase in dosage did not cause significant uptake. Maximum removal of total hardness (72%) was achieved at 20g/l dosage. Increase in adsorbent dosage from 25g/l to 30g/l resulted in decrease in removal efficiency. This may be due to the presence of $CaCO_3$, the main constituent of snail shell which increased the Ca^{2+} hardness of the treated effluent as well as the ion-exchange capacity of snail shell.

Figure 1: Effect of adsorbent dosage on the removal of contaminants

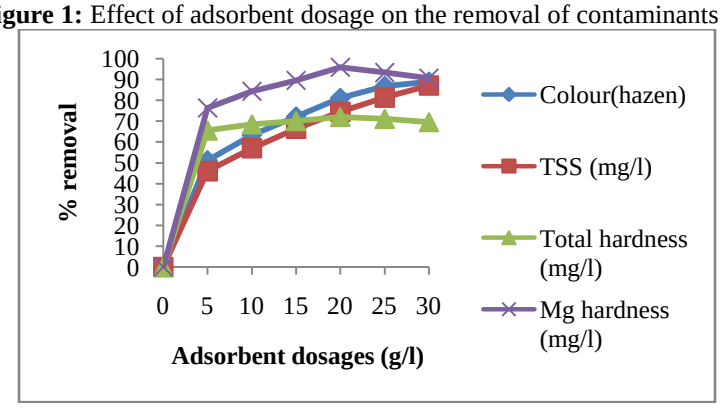


Figure 2: Effect of adsorbent dosage on Ph

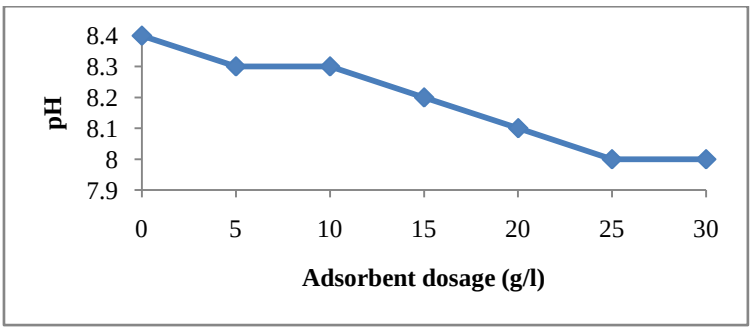


Figure 3 shows the effect of varying dosages on the concentration of other contaminants in treated effluent.

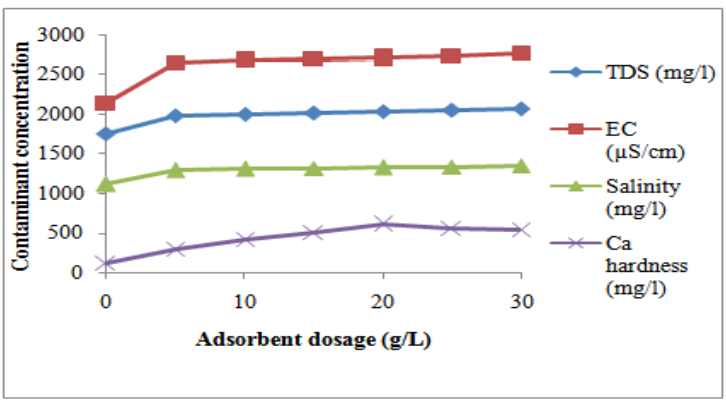
The Electrical Conductivity (EC), Total Dissolved Solids (TDS), Salinity and Ca hardness of treated effluent increased proportionally with increasing adsorbent dosages. This is due to the partial dissolution and possible interaction between the snail shell activated carbon and constituents of treated wastewater.

The increase in Conductivity and Ca hardness can be attributed to the calcium-rich nature of snail shell. The major composition of snail shell is calcium carbonate (97.5%), however, there are other elements like calcium phosphate, calcium silicate, magnesium carbonate, magnesium oxide, iron oxide, manganese oxide and other organic substances [25]. Thus, as the adsorbent doses increased, the total ions that produced the electric current also increased.

The increase in salinity can be attributed to the presence of residual calcium phosphate salt, $Ca_3(PO_4)_2$ from the chemical activation of $CaCO_3$ rich snail shell with ortho-phosphoric acid, H_3PO_4 .

The increase in EC can be attributed to major positive ions species like Ca^{2+}, Mg^{2+}, Fe^{2+} and Mn^{2+} found in snail shell and residual H^+ ions from acid activation of precursor. Other minor contributors to conductivity are CO_3^{2-} and PO_4^{3-} derived from snail shell and phosphoric acid respectively.

Figure 3: Adsorbent dose effect on other contaminants



3.2 Equilibrium Study

In this study, adsorption data was described by adsorption isotherms of Freundlich and Langmuir. These isotherms relate adsorbate uptake per unit mass of adsorbent, q_e, to the equilibrium adsorbate concentration in the bulk fluid phase C_e.

3.2.1 The Freundlich Isotherm

The Freundlich isotherm model is an empirical relationship describing the adsorption of solutes from a liquid to a solid surface and assumes that different sites with several adsorption energies are involved. Freundlich adsorption isotherm is the relationship between the amounts of contaminant adsorbed per unit mass of adsorbent, q_e and the concentration of the contaminant at equilibrium, C_e.

$$q_e = k_f C_e^{1/n} \quad (3)$$

The logarithmic form of the equation becomes:

$$\log q_e = \log K_f + \frac{1}{n}\log C_e \quad (4)$$

where, K_f and n are the Freundlich constants, the characteristics of the system. K_f and n are the indicators of the adsorption capacity and adsorption intensity respectively. The ability of Freundlich model to fit the experimental data was examined. For this case, the plot of log q_e vs. log C_e was employed to generate the intercept value of K_f and the slope of n.

From Figures 4 and 5, the values of constant *1/n* were 0.751 and 0.743 for colour and TSS removal respectively. The high R^2 suggests the applicability of the Freundlich isotherm in describing the adsorption process. The constant 1/n classifies the isotherm to be irreversible (1/n=0), favourable (0< 1/n <1) and unfavourable (1/n>1) [3]. The Freundlich constants K_f and n were found to be 0.668 and 1.332 for color and 0.916 and 1.346 for TSS respectively as shown in Tables 2 and 3. The magnitudes of K_f and n show easy separation of dyestuff and suspended solids from the textile wastewater onto the prepared activated snail shell adsorbent surface and also indicate favourable adsorption. From Tables 2 and 3, the n value is high enough for separation. The Freundlich isotherm fitted well with the correlation coefficient of 0.932 and 0.874 for color and TSS respectively.

Figure 4: Freundlich adsorption isotherm for Color removal

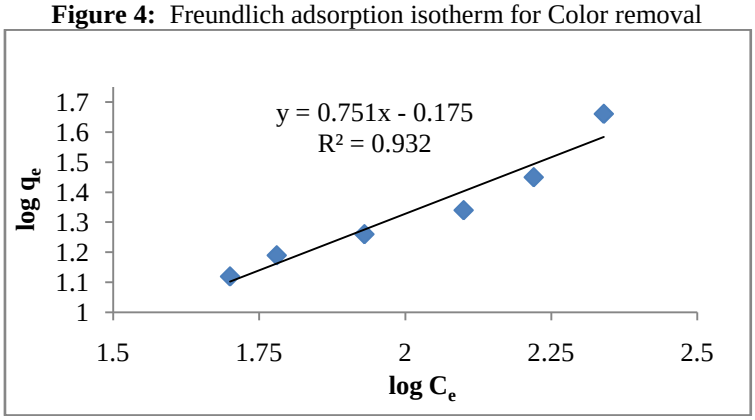
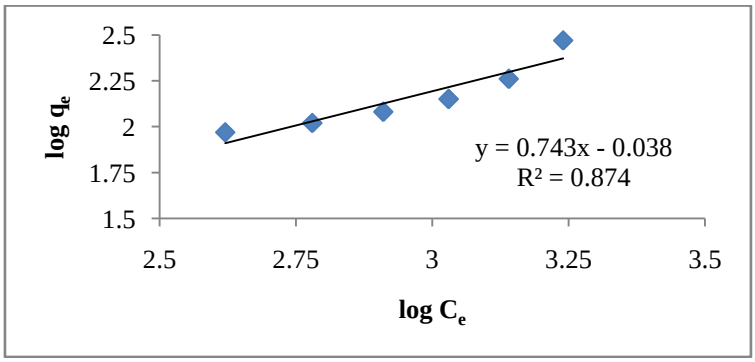


Figure 5: Freundlich adsorption isotherm for TSS removal



3.2.2 The Langmuir isotherm

The Langmuir model is based on the assumption that maximum adsorption occurs when a saturated monolayer of solute molecules is present on the adsorbent surface, the energy of adsorption is constant and there is no migration of adsorbate molecules in the surface plane. The Langmuir isotherm is given by:

$$q_e = \frac{q_m K_L C_e}{1+K_L C_e} \quad (5)$$

The constants in the Langmuir isotherm were determined by plotting ($1/q_e$) versus ($1/ C_e$) to conform with the equation rewritten as:

$$\frac{1}{q_e} = \frac{1}{q_m} + \frac{1}{q_m K_L}\frac{1}{C_e} \quad (6)$$

Where, q_m and K_L are the Langmuir constants, representing the maximum adsorption capacity for the solid phase loading and the energy constant related to the heat of adsorption respectively. The essential characteristics of Langmuir adsorption isotherm can be expressed in terms of a dimensionless constant called the separation factor or equilibrium parameter R_L [18] which describes the type of isotherm and is defined by:

$$R_L = \frac{1}{(1+K_L C_o)} \quad (7)$$

Where, K_L is Langmuir Isotherm Constant; C_o is the highest initial concentration of contaminant.
The value of R_L indicates the type of the isotherm to be either unfavorable ($R_L > 1$), linear ($R_L = 1$), favourable ($0 < R_L < 1$) or irreversible ($R_L = 0$) [28].

Figures 6 and 7 show that the data fits the Langmuir isotherm well with R^2 values of 0.938 and 0.851 for color and TSS removal respectively. This is in agreement with the findings of [14] for removal of Methylene Blue dye from solution with activated snail shell adsorbent. The values of q_m and K_L were found to be 65.3 hu/g and 0.005 L/hu for color and 349.4 mg/g and 7.87 L/mg for TSS respectively. The R_L values were 0.308 and 0.284 for color and TSS removal respectively. This suggests that the adsorption of color and TSS onto prepared snail shell adsorbent is favourable.

Figure 6: Langmuir adsorption isotherm for Color removal

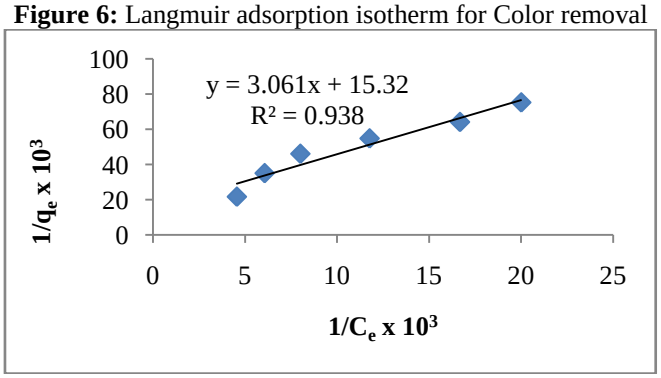


Figure 7: Langmuir adsorption isotherm for TSS removal

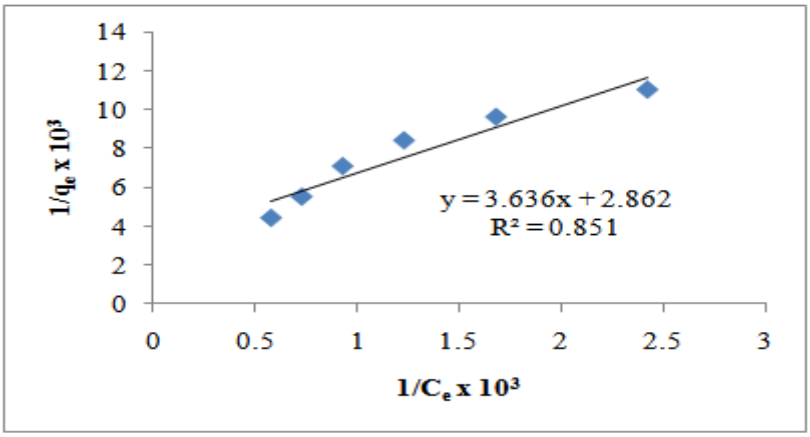


Table 2: Isotherm Model Constants and Correlation Coefficients for Adsorption of Color with Snail Shell AC

Freundlich Isotherm			*Langmuir Isotherm*			
K_f	*n*	R^2	q_m *(hu/g)*	K_L *(L/hu)*	R^2	R_L
0.668	1.332	0.932	65.3	0.005	0.938	0.308

Table 3: Isotherm Model Constants and Correlation Coefficients for Adsorption of TSS with Snail Shell AC

Freundlich Isotherm			*Langmuir Isotherm*			
K_f	*n*	R^2	q_m *(mg/g)*	K_L *(L/mg)*	R^2	R_L
0.916	1.346	0.874	349.4	7.87	0.851	0.284

IV. Conclusion

The study has shown that activated carbon derived from African Giant Snail shell is a good adsorbent for Color, Total Suspended Solids (TSS) and Mg hardness in textile wastewater. A reduction of 88.9% and 87.1% at optimum dosage of 30g/l was achieved for Colour and TSS respectively. At optimum dosage of 20g/l, about 72% of Total hardness was removed. The TDS, Electrical Conductivity, Salinity and Ca hardness increased with carbon dosage, this being attributed to the high calcium content of snail shell. The adsorption of Color and TSS onto prepared snail shell adsorbent was well described by Freundlich and Langmuir isotherms with high R^2 values. The outcome of the study proves that snail shell derived activated carbon is an effective biosorbent for decolourization and detoxification of textile wastewater in addition to their low-cost and environment-friendly benefits.

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Dr. Amal Kumar Ghosh. " Life and Environment – Sequence and Pre conditions." IOSR Journal of Environmental Science, Toxicology and Food Technology (IOSR-JESTFT) 12.4 (2018): 26-33.