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Removal of Heavy Metals from Water Accompanied with Oil and Wastewater, Using Natural Dyes and Chemical Compounds.in Elwahate Jalu, Libya

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الملخص:

تتميز النباتات سواء كانت الأوراق او الثمار او اللحاء بكم هائل من المركبات العضوية التي يمكن استخدامها لتكوين متراكبات مع العناصر الثقيلة والسامة الملوثة للبيئة باعتبار هذه المستخلصات النباتية مركبات صديقة للبيئة وتم في هذا العمل عمل مستخلصات من نباتات القرنفل والقرفة والمريمية وأوراق النعناع والمارينغا والجزر والملفوف والبنجر والبرتقال والريحان وأظهرت هذه المستخلصات قدرات مختلفة على ترسيب العديد من الأيونات الثقيلة الذائبة في الماء مثل عناصر النحاس والكادميوم والنيكل والفضة والرصاص والحديد والكروم والكالسيوم والخارصين والمنجنيز. كما أظهرت العديد من المستخلصات مثل القرنفل والقرفة والنعناع والمرمرية وغيرها قدرات عالية على ترسيب العناصر الثقيلة داخل محاليلها وأمكن تطبيق ذلك على مياه الصرف والمياه المصاحبة للنفط ومياه الصرف الصناعي وأظهرت أيضا قدرة عالية على ترسيب هذه الأيونات ومتابعة عملية المعالجة في جميع حالاتها باستخدام جهاز الاسبكتروفوتومتر.

الكلمات المفتاحية:

المركبات العضوية، الفواكه، العناصر الثقيلة، الواحات جالو، ليبيا.

Abstract

Plants, whether leaves, fruits or bark, are characterized by an enormous number of organic compounds that can be used to form complexes with heavy and toxic elements that polluting the environment as these plant extracts are environmentally friendly compounds. In this paper, extracts from plants and the fruits like cloves, cinnamon bark, beet fruits, carrots, oranges, mint leaves, basil, Maringa, red cabbage, and sage have been performed. These extracts have shown different capabilities to precipitate many heavy ions dissolved in water, such as copper, cadmium, nickel, silver, lead, iron, chromium, calcium, zinc and manganese. Precipitation of heavy elements in their solutions. This could be applied to wastewater and water accompanied with oil and industrial waste water. Showing a high ability to precipitate these ions and follow the treatment process in all of its cases using a spectrophotometer.

Keywords: organic compounds; fruits; heavy metals; Elwahate Jalu-Libya.

1. INTRODUCTION

The present research, deals with organic compounds that can be form complexes with heavy and toxic elements that polluting the environment as these plant extracts are environmentally friendly compounds. Complementary insights regarding heavy metal behavior and phyto-availability in soils and sediments have been proposed by selective single extraction procedures [1]. One of the prime challenges to the present world is the removal of organic dyes from wastewater by innovative effluent treatment plant, which can truly clean the wastewater without leaving any fragments of dye species and not generating secondary waste [2]. The naturally available carbohydrate polymeric biodegradable adsorbent of wheat flour (WF) for the encapsulation of cationic Rhodamine B (RB) dye with high sensitivity and selectivity, was investigated [3]. Using the natural clay (Sale-Morocco) for adsorption of heavy metals (Cu, Co, Ni and Pb) as a potential adsorbent in the treatment synthetic wastewater [4]. Using wastewater treatment by micro-plastics pollution in aquatic ecosystems is of great concern; however, systemic investigations are still lacking in freshwater wetland systems [5].

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Hydrothermal carbonization (HTC) at different conditions used for the distribution and risk as well as leaching toxicity of heavy metals in textile sludge [6]. Remove heavy metals by bio-mineralization method in river sediments by the *Bacillus subtilis.* Optimal content of *Bacillus subtilis* powder and organophosphate monoester was 20 g and 0.2 mol in 1 L of bacterial solution, respectively [7]. An innovative strategy based on the gelation process of Sodium Alginate (SA) was proposed for synchronous treatment of composite pollutants [8]. A drinking water treatment residual (DWTR)-based granule (DBG) substrate was developed herein by pyrolyzing and granulating the DWTR with bentonite and corncob [9.] Heavy metals (Cu, Pb, Zn, Ni, Cr, and Cd) were removed from sludge via joint treatment with ethylene diamine tetra acetic acid (EDTA) and three organic acids (citric acid, glutamic acid, or aspartic acid) at optimal EDTA-acid concentration ratios of 1:1, 1:2 and 2:1, respectively [10].

2. METHODS

2.1. Chemicals and Used equipment's

100 beakers of 100 ml capacity - filter papers - funnels - cylinder tester - cup - conical flasks – spectrophotometer (Jenway 6300 spectrophotometer) - sensitive balance - standard 100 ml beakers

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2.2. Preparation of solutions

100 ml of solution of 0.1 molar are prepared from the following materials:1/ Cu $SO_4.5H_2O$ (2.4968g), 2/ Ca $(NO_3)_2$. $4H_2O$ (2.6313g), 3/ $AgNO_3$ (1.9687g), 4/ $NiCl_2.6H_2O$ (2.7371g), 5/Fe $SO_4.7H_2O$ (2.27g) 6/ Cr $(NO_3)_3.9H_2O$ (4.006g), 7/ Cd $(NO_3)_3.4H_2O$ (3.0848g), 8/ $(CH_3COO)_2Pb$ $3H_2O$ (3.7933g), 9/ $ZnSO_4.7H_2O$ (1.815g) and 10/ Mn SO_4. H_2O (1.9602g)

2.3. Preparation of plant extracts

In 100 ml distilled water boil 5g of the leaves of some plants or fruits, such as Cloves, Cinnamon, Sage, Mint, Maringa, Carrots, Cabbage, Beets, Orange , and Basil, for three minutes, about 100 ml of their extractions and juices with distilled water are filtered.

2.4. The method of work

The absorbance of each of the ion solutions and the extracts is measured separately, then about 10 ml of each type of plant extract is added to 10 ml of the solutions of heavy elements and left for 24 hours.

The application to treatment the wastewater, water accompanied with oil and battery waste water by difference weight method (add 10 ml of extract to 10ml of 0.1M of heavy metal ions and weight the precipitate then add10 ml of extract to 10ml of 0.1M of heavy metal ions and 10 ml of waste water. Calculating the difference between two weights, gives the total heavy metals precipitate in wastewater and compare the data with direct method of precipitation of heavy metals by extractions directly.

(Table 1).

Table 1. The measurements of milligram weight of precipitate and metal ion solutions precipitation the different samples from 1/wastewater, 2/water accompanied with oil and 3/battery wastewater

extraction only	1mg	2 mg	3 mg
Cloves	37	346	60
Cinnamon	29	509	143
Sage	9	355	60
Mint	77	369	96
Maringa	20	290	60
Carrots	54	338	76
Cabbage	25	358	159
Beets	51	380	66
Orange	36	350	48
Basil	52	370	20

3. RESULTS

A 0.1 mol of copper sulfate solution was treated by plant extracts of clove, cinnamon, sage, mint, Maringa, carrot, cabbage, beet, orange and basil (Fig. 1). These extracts showed different abilities to precipitate many heavy ions dissolved in water from copper in varying levels, and the largest precipitate of copper was the extract of carrots, then basil and sage, due to its may be formed a complex with copper ion precipitate as shown in figure 1 which shows that there are three higher weight precipitate of copper complexes which may be formed with Carrots, Basil and Sage.

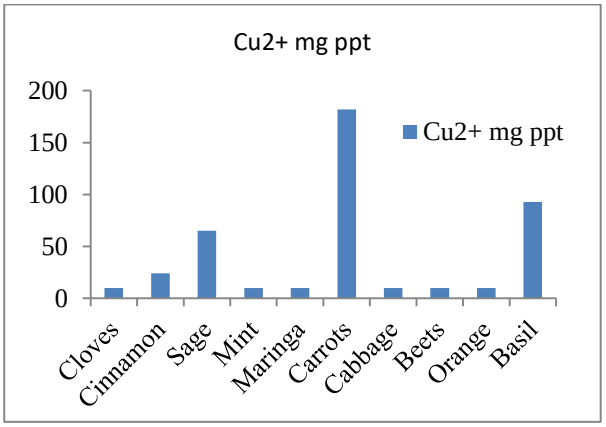


Figure 1. Shows the weight precipitate in milligrams of copper complexes which may be formed by adding 10 ml of extract to 10 ml of 0.1M Cu $SO_4.5H_2O$.

A 0.1 mol of calcium nitrate Ca $(NO_3)_2$. $4H_2O$. solution was treated by plant extracts of clove, cinnamon, sage, mint, Maringa, carrot, cabbage, beet, orange and basil (Fig.2). These extracts showed different abilities to precipitate many heavy ions dissolved in water from copper in varying levels, and the largest precipitate of Calcium was the extract of clove, and sage, due to its may be formed a complex with Calcium ion precipitate as shown in figure 1, which shows that there are two higher weight precipitate of Calcium complexes which may be formed with Cloves and Sage.

A 0.1 mol of silver nitrate $AgNO_3$ solution was treated by plant extracts of clove, cinnamon, sage, mint, Maringa, carrot, cabbage, beet, orange and basil (Fig. 3). These extracts showed different abilities to precipitate many heavy ions dissolved in water from copper in varying levels, and the largest precipitate of silver was the extract of clove, cabbage, orange and basil, due to its may be formed a complex with silver ion precipitate as shown in figure 3, which shows that there are four higher weight precipitate of silver complexes which may be formed with Cloves, Cabbage, Orange and Basil.

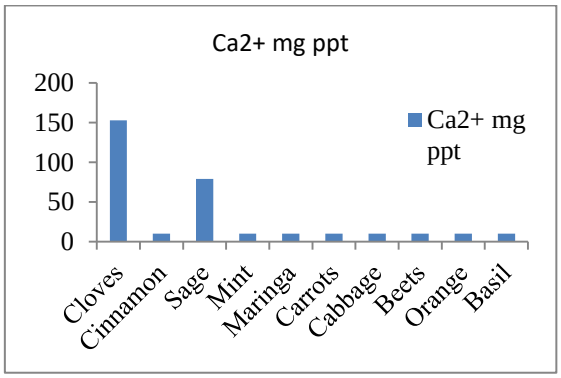


Figure 2. Shows the weight precipitate in milligrams of Calcium complexes which may be formed by adding 10 ml of extract to 10 ml of 0.1M Ca $(NO_3)_2$. $4H_2O$.

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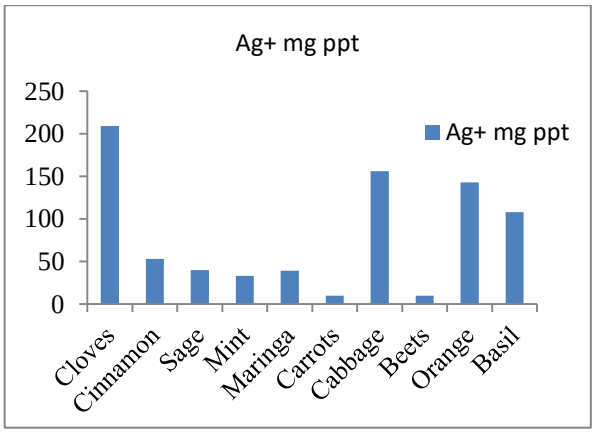


Figure 3. Shows the weight precipitate in milligrams of silver complexes which may be formed by adding 10 ml of extract to 10 ml of 0.1M $AgNO_3$.

A 0.1 mol of nickel chloride $NiCl_2.6H_2O$. solution was treated by plant extracts of clove, cinnamon, sage, mint, maringa, carrot, cabbage, beet, orange and basil (Fig. 4). These extracts showed different abilities to precipitate many heavy ions dissolved in water from copper in varying levels, and the largest precipitate of nickel was the extract of clove, sage, and mint, due to its may be formed a complex with nickel ion precipitate as shown in figure 4, which shows that there are three higher weight precipitate of Nickel complexes which may be formed with Cloves, Sage and Mint.

A 0.1 mol of ferrous sulphate Fe $SO_4.7H_2O$. solution was treated by plant extracts of clove, cinnamon, sage, mint, Maringa, carrot, cabbage, beet, orange and basil (Fig. 5). These extracts showed different abilities to precipitate many heavy ions dissolved in water from copper in varying levels, and the largest precipitate of ferrous was the extract of clove, cinnamon, sage, mint, carrot, cabbage, beet, and basil. due to its may be formed a complex with ferrous ion precipitate as shown in figure 5, which shows that there are eight higher weight precipitate of Ferrous complexes which may be formed with Cloves, Cinnamon, Sage, Mint, Carrots, Cabbage, Beets and Basil.

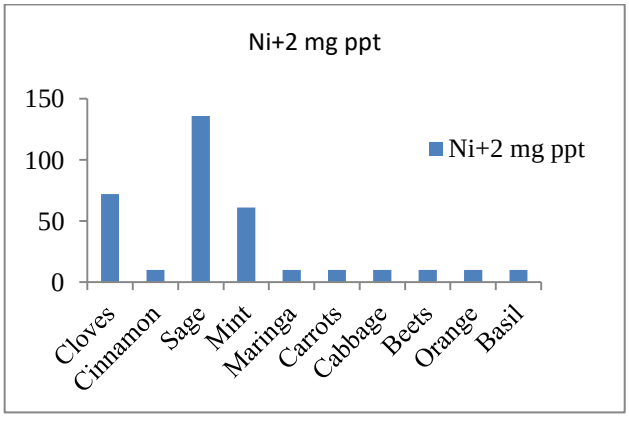


Figure 4. Shows the weight precipitate in milligrams of Nickel complexes which may be formed by adding 10 ml of extract to 10 ml of 0.1M $NiCl_2.6H_2O$.

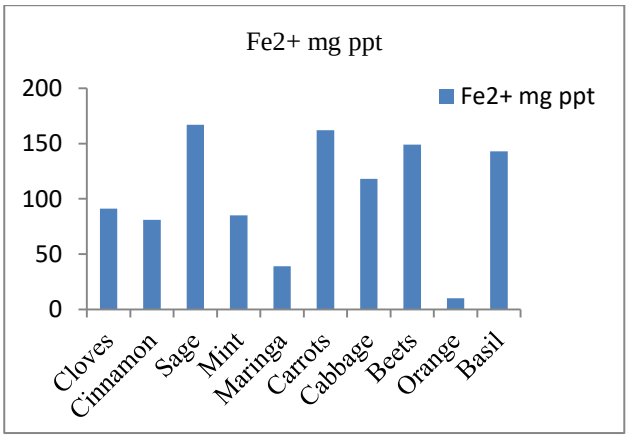


Figure 5. Shows the weight precipitate in milligrams of Ferrous complexes which may be formed by adding 10 ml of extract to 10 ml of 0.1M Fe $SO_4.7H_2O$.

A 0.1 mol of chromium nitrate Cr $(NO_3)_3.9H_2O$. solution was treated by plant extracts of clove, cinnamon, sage, mint, Maringa, carrot, cabbage, beet, orange and basil (Fig. 6). These extracts showed different abilities to precipitate many heavy ions dissolved in water from chromium ion in varying levels, and the largest precipitate of chromium was the extract of clove, sage, and mint, due to its may be formed a complex with chromium ion precipitate as shown in figure 6, which shows that there are three higher weight precipitate of Chromium complexes which may be formed with Cloves, Sage and Mint.

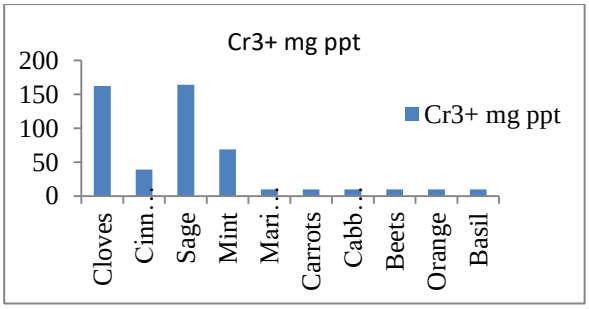


Figure 6. Shows the weight precipitate in milligrams of Chromium complexes which may be formed by adding 10 ml of extract to 10 ml of 0.1M Cr $(NO_3)_3.9H_2O$.

A 0.1 mol of cadmium nitrate Cd $(NO_3)_3.4H_2O$. solution was treated by plant extracts of clove, cinnamon, sage, mint, maringa, carrot, cabbage, beet, orange and basil (Fig. 7). These extracts showed different abilities to precipitate many heavy ions dissolved in water from cadmium ion in varying levels, and the largest precipitate of cadmium was the extract of clove, sage, and mint, due to its may be formed a complex with cadmium ion precipitate as same as chromium as shown in figure 7, which shows that there are three higher weight precipitate of Cadmium complexes which may be formed with Cloves, Sage and Mint.

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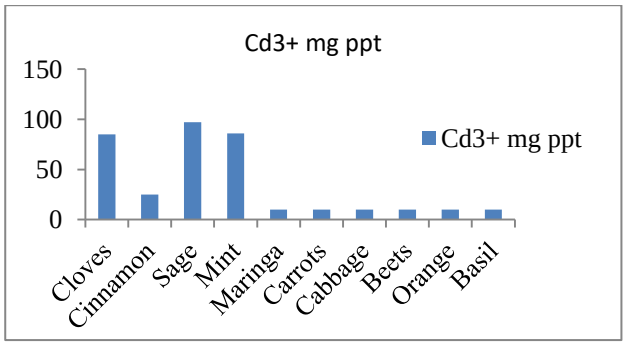


Figure 7. Shows the weight precipitate in milligrams of Cadmium complexes which may be formed by adding 10 ml of extract to 10 ml of 0.1M Cd $(NO_3)_3.4H_2O$.

A 0.1 mol of lead acetate $CH_3COO)_2Pb\ 3H_2O$.solution was treated by plant extracts of clove, cinnamon, sage, mint, maringa, carrot, cabbage, beet, orange and basil (Fig. 8). These extracts showed different abilities to precipitate many heavy ions dissolved in water from lead in varying levels, and the largest precipitate of lead was the extract of clove, sage, carrot, cabbage, beet, and orange, due to its may be formed a complex with lead ion precipitate as shown in figure 8, which shows that there are seven higher weight precipitate of Lead complexes which may be formed with Cloves, Sage, Mint, Maringa, Carrots, Cabbage, Beets and Orange.

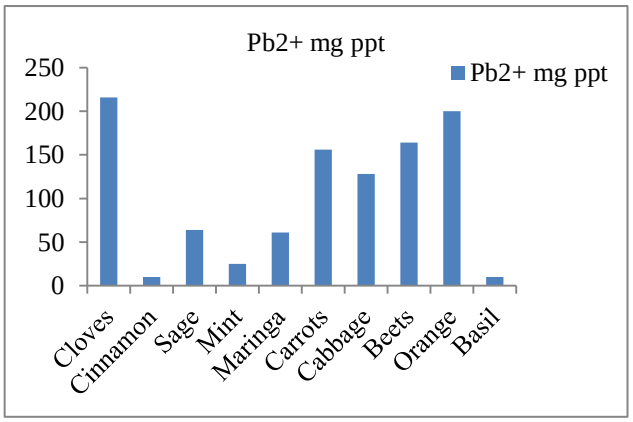


Figure 8. Shows the weight precipitate in milligrams of Lead complexes which may be formed by adding 10 ml of extract to 10 ml of 0.1M $(CH_3COO)_2Pb\ 3H_2O$.

A 0.1 mol of zinc sulphate $ZnSO_4.\ 7H_2O$.solution was treated by plant extracts of clove, cinnamon, sage, mint, Maringa, carrot, cabbage, beet, orange and basil (Fig.9). These extracts showed different abilities to precipitate many heavy ions dissolved in water from zinc in varying levels, and the largest precipitate of zinc was the extract of clove, carrot, beet, and basil, due to its may be formed a complex with zinc ion precipitate as shown in figure 9, which shows that there are five higher weight precipitate of Zinc complexes which may be formed with Cloves, Cinnamon, Carrots, Beets and Basil.

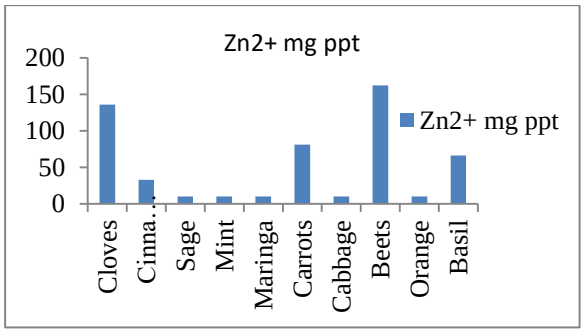


Figure 9. Shows the weight precipitate in milligrams of Zinc complexes which may be formed by adding 10 ml of extract to 10 ml of 0.1M $ZnSO_4.7H_2O$.

A 0.1 mol of manganese sulphate Mn $SO_4.\ H_2O$. solution was treated by plant extracts of clove, cinnamon, sage, mint, Maringa, carrot, cabbage, beet, orange and basil (Fig. 10). These extracts showed different abilities to precipitate many heavy ions dissolved in water from manganese in varying levels, and the largest precipitate of manganese was the extract of clove, cinnamon, and sage, due to its may be formed a complex with manganese ion precipitate as shown in figure 10, which shows that there are three higher weight precipitate of Manganese complexes which may be formed with Cloves, Cinnamon and Sage.

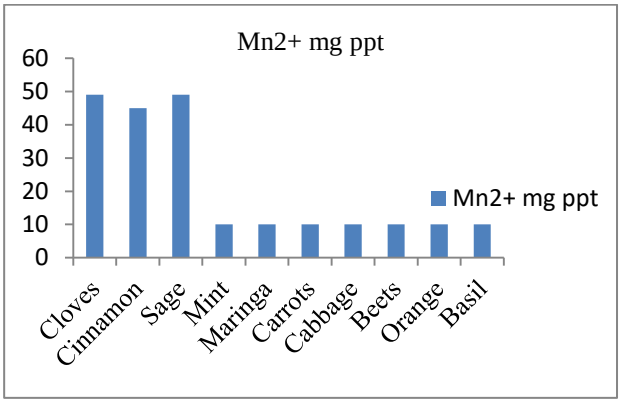


Figure 10. Shows the weight precipitate in milligrams of Manganese complexes which may be formed by adding 10 ml of extract to 10 ml of 0.1M Mn $SO_4.\ H_2O$.

4. DISCUSSION

Waste water contain many of heavy metal ions dissolved in water It is toxic and dangerous to humans, animals and plants, so it must be precipitate of by treating it with cheap and available materials in the surrounding environment, such as the following plant extracts clove, cinnamon, sage, mint, Maringa, carrot, cabbage, beet, orange and basil. Those extracts showed varying abilities in precipitating and treating those heavy ions such as cloves and cinnamon as show in (Figs. 11, 12). By difference weight method and direct weight method. Figure 11 explains the ability of differences of the extractions to precipitate the heavy metal ions in waste water and the highest extraction is Cloves using the difference weight method. Figure 12 explains the ability of differences of the extractions to precipitate the heavy metal ions in wastewater and the highest extraction is Mint using the direct weight method.

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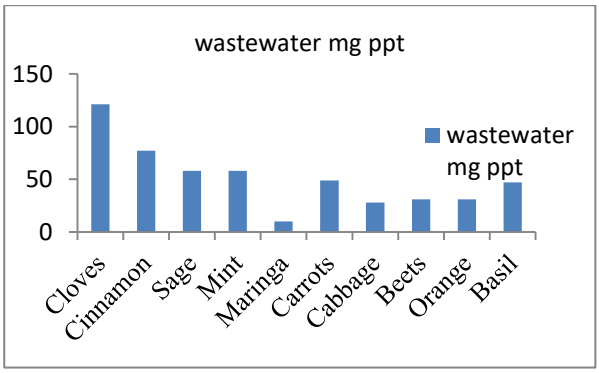


Figure 11. Shows the weight of metal ions precipitate in milligrams of wastewater with natural extractions by difference weight method.

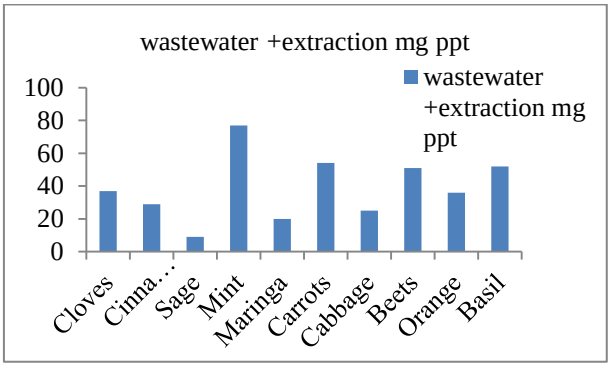


Figure 12. Shows the weight of metal ions precipitate in milligrams of wastewater with natural extractions by direct weight method.

water accompanied with oil contain many of heavy metal ions dissolved in water It is toxic and dangerous to humans, animals and plants, so it must be precipitate of by treating it with cheap and available materials in the surrounding environment, such as the following plant extracts clove, cinnamon, sage, mint, Maringa, carrot, cabbage, beet, orange and basil. Those extracts showed varying abilities in precipitating and treating those heavy ions such as cloves and sage as show in (Figs. 13, 14). By difference weight method and direct weight method. It is noted that the water accompanied with oil contains huge amounts of heavy metal ions, which makes it very dangerous to the environment. Which is approximately four times the ions present in waste water. Figure 13 explains the ability of differences of the extractions to precipitate the heavy metal ions in water accompanied with oil and the highest extraction is Sage using the difference weight method. Figure 14, explains the ability of differences of the extractions to precipitate the heavy metal ions in water accompanied with oil and the highest extraction is Cinnamon using the direct weight method.

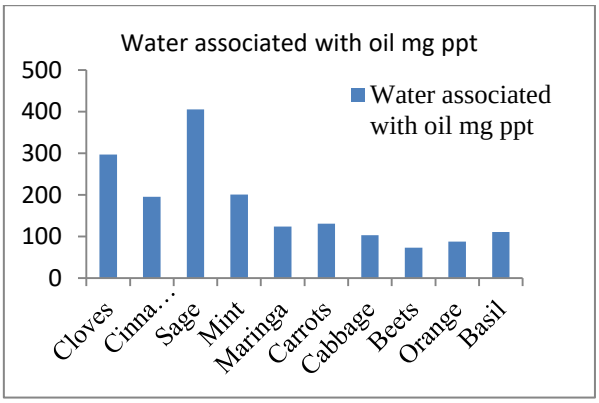


Figure 13. Shows the weight of metal ions precipitate in milligrams of water accompanied with oil with natural extractions by difference weight method.

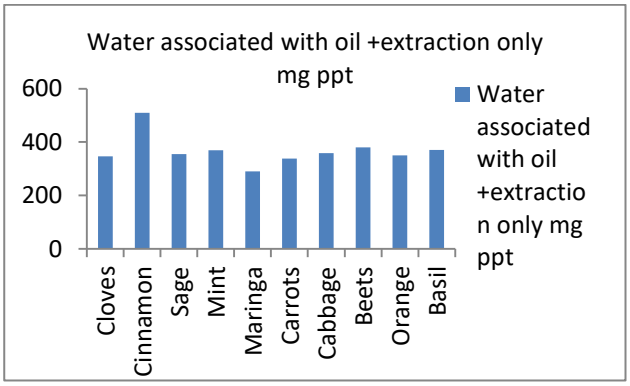


Figure 14. Shows the weight of metal ions precipitate in milligrams of water accompanied with oil with natural extractions by direct weight method.

Batteries water, when discharged and thrown into waste water, contains many heavy metal ions, the most dangerous and most dangerous of which are lead ions. Treating these ions with plant extracts showed a superior ability to precipitate these ions, making it a quick, cheap and highly efficient solution in treating heavy ions in all types of water, especially battery water (Figs. 15, 16). By difference weight method and direct weight method, Figure 15, explains the ability of differences of the extractions to precipitate the heavy metal ions in water accompanied with oil and the highest extraction is Carrots using the difference weight method. Figure 16 explain the ability of differences of the extractions to precipitate the heavy metal ions in water accompanied with oil and the highest extraction is Red Cabbage using the direct weight method.

In this part of the paper, each of the ten extracts was studied with the ions of the ten heavy metals under study, and the efficiency and ability of each extract to precipitate ions were studied, and the deposition efficiency was confirmed by measuring the high wavelength of the filter (Figs. 17-26).

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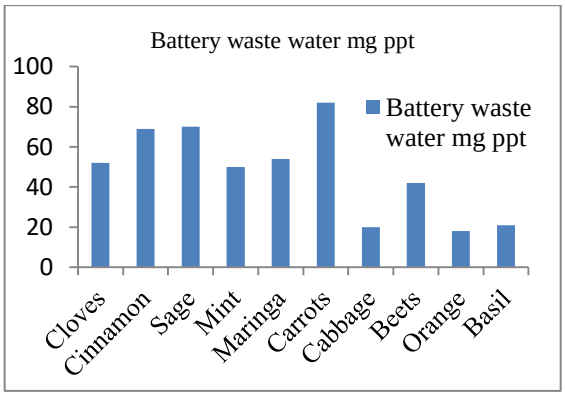


Figure 15. Shows the weight of metal ions precipitate in milligrams of battery wastewater with natural extractions by difference weight method.

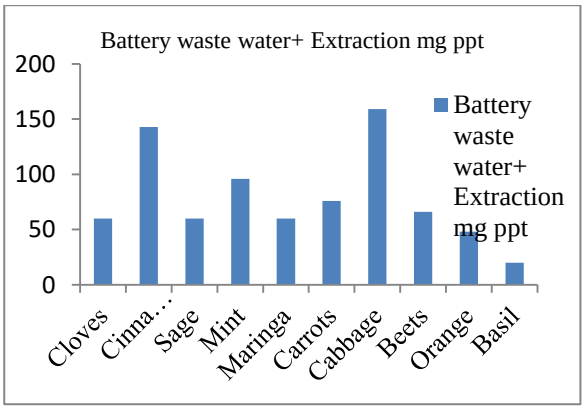


Figure 16. Shows the weight of metal ions precipitate in milligrams of battery wastewater with natural extractions by direct weight method.

The sedimentation efficiency of the extracts is high at these ions. Figure 17 explains the weight in milligrams of precipitate of heavy metals with the Cloves and the high metal ions precipitated are Ag^{+}, Cr^{3+} and Pb^{2+}. The constant $\lambda_{max.}$ of filtrate indicate that there are not metal ions or complexes in solution. Figure 18 explains the weight in milligrams of precipitate of heavy metals with the Cinnamon and the high metal ions precipitated are Ag^{+}, Fe^{2+}, Cr^{3+} Cd^{3+} and Zn^{2+}. The constant $\lambda_{max.}$ of filtrate indicate that there are not metal ions or complexes in solution. Figure 19 explains the weight in milligrams of precipitate of heavy metals with the Sage and the high metal ions precipitated are Ca^{2+}, Fe^{2+}, Cr^{3+} Cd^{3+} and Mn^{2+}. The constant $\lambda_{max.}$ of filtrate indicate that there are not metal ions or complexes in solution. Figure 20 explains the weight in milligrams of precipitate of heavy metals with the Mint and the high metal ions precipitated are Ni^{2+}, Fe^{2+}, Cr^{3+} Cd^{3+} and Pb^{2+}. The constant $\lambda_{max.}$ of filtrate indicate that there are not metal ions or complexes in solution. Figure 21 explains the weight in milligrams of precipitate of heavy metals with the Maringa and the high metal ions precipitated are Ag^{+}, Fe^{2+} and Pb^{2+}. The constant $\lambda_{max.}$ of filtrate indicate that there are not metal ions or complexes in solution. Figure 22 explains the weight in milligrams of precipitate of heavy metals with the Carrots and the high metal ions precipitated are Cu^{2+}, Fe^{2+} and Pb^{2+}. The constant $\lambda_{max.}$ of filtrate indicate that there are not metal ions or complexes in solution. Figure 23 explains the weight in milligrams of precipitate of heavy metals with the Red Cabbage and the high metal ions precipitated are Ag^{+}, Fe^{2+} and Pb^{2+}. The constant $\lambda_{max.}$ of filtrate indicate that there are not metal ions or complexes in solution. Figure 24 explains the weight in milligrams of precipitate of heavy metals with the Beets and the high metal ions precipitated are Fe^{2+}, Zn^{2+} and Pb^{2+}. The constant $\lambda_{max.}$ of filtrate indicate that there are not metal ions or complexes in solution. Figure 25 explains the weight in milligrams of precipitate of heavy metals with the Orange and the high metal ions precipitated are Ag^{+} and Pb^{2+}. The constant $\lambda_{max.}$ of filtrate indicate that there are not metal ions or complexes in solution. Figure 26 explains the weight in milligrams of precipitate of heavy metals with the Beets and the high metal ions precipitated are Fe^{2+}, Ag^{+} and Zn^{2+}. The constant $\lambda_{max.}$ of filtrate indicate that there are not metal ions or complexes in solution.

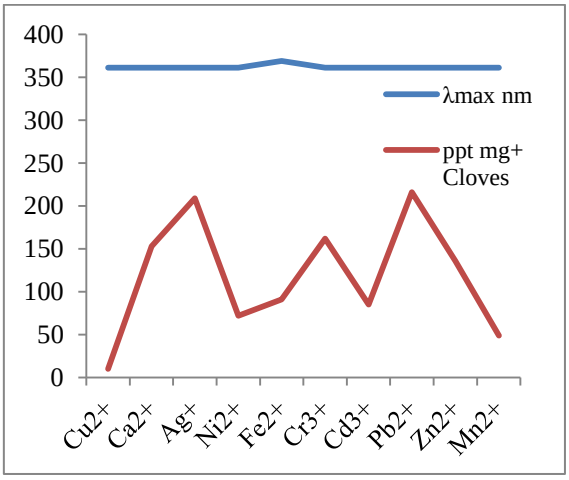


Figure 17. Shows the weight of metal ions precipitate in milligrams with Cloves and corresponding maximum wavelength $\lambda_{max.}$ of filtrate.

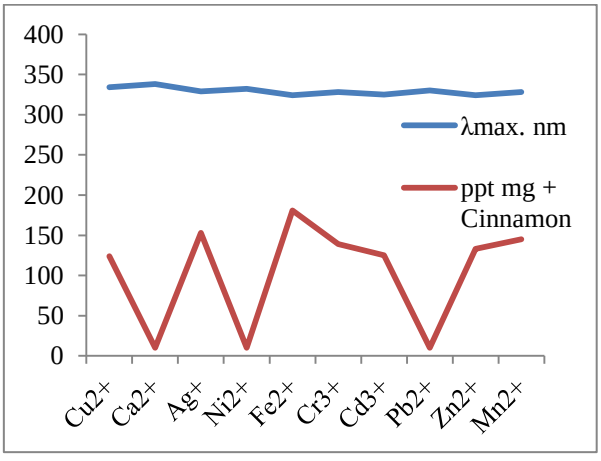


Figure 18. Shows the weight of metal ions precipitate in milligrams with Cinnamon and corresponding maximum wavelength $\lambda_{max.}$ of filtrate.

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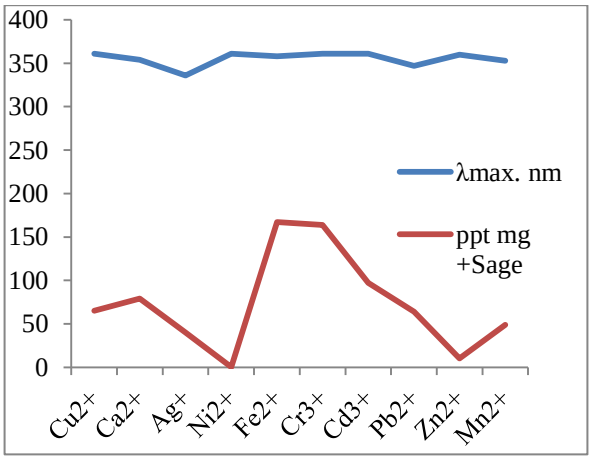


Figure 19. Show the weight of metal ions precipitate in milligrams with Sage and corresponding maximum wavelength $\lambda_{max.}$ of filtrate.

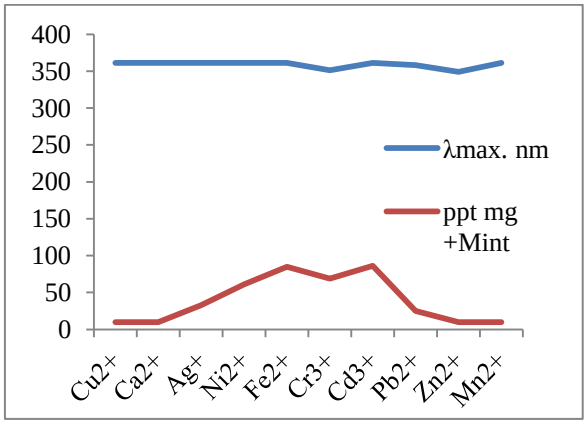


Figure 20. Shows the weight of metal ions precipitate in milligrams with Mint and corresponding maximum wavelength $\lambda_{max.}$ of filtrate.

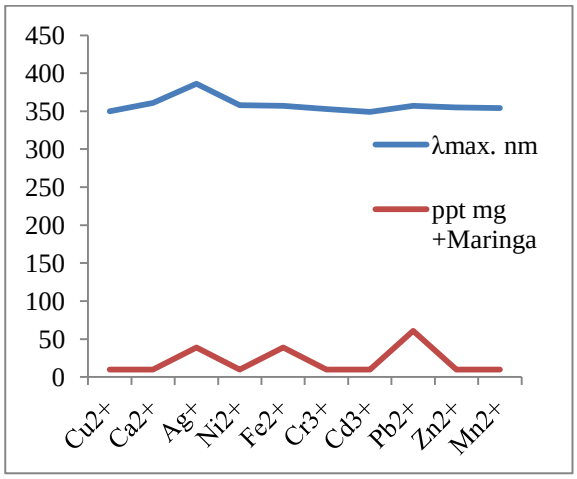


Figure 21. Shows the weight of metal ions precipitate in milligrams with Maringa and corresponding maximum wavelength $\lambda_{max.}$ of filtrate.

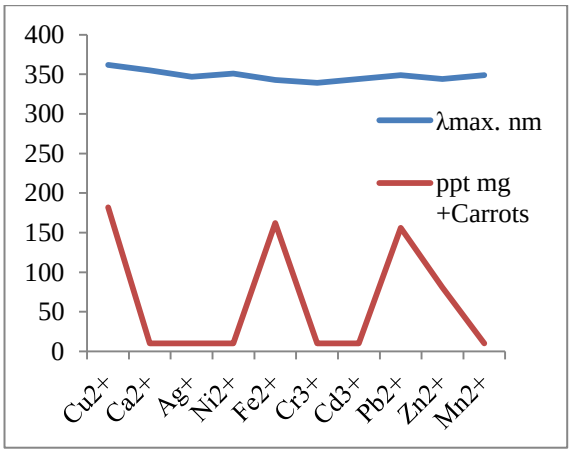


Figure 22. Shows the weight of metal ions precipitate in milligrams with Carrots and corresponding maximum wavelength $\lambda_{max.}$ of filtrate.

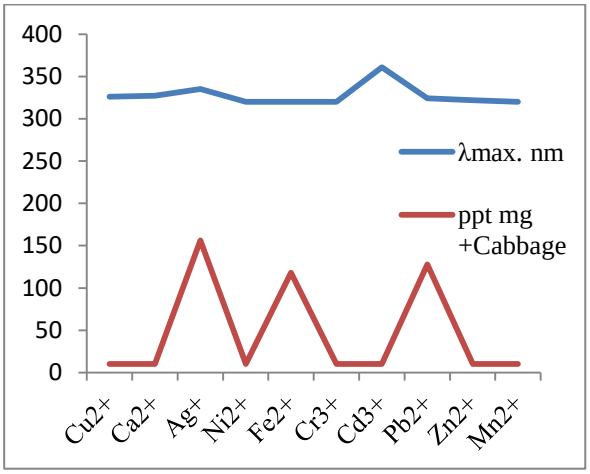


Figure 23. Shows the weight of metal ions precipitate in milligrams with Red Cabbage and corresponding maximum wavelength $\lambda_{max.}$ of filtrate.

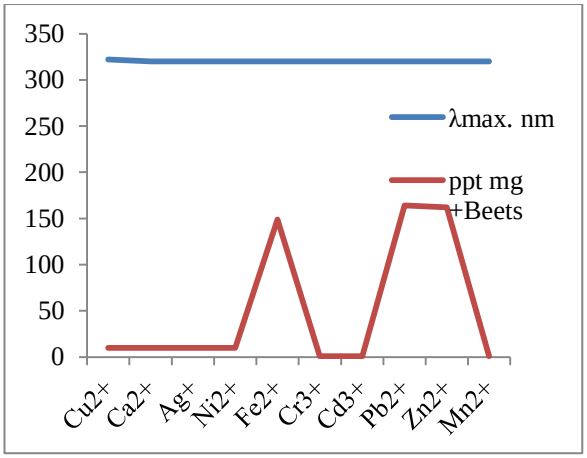


Figure 24. Shows the weight of metal ions precipitate in milligrams with Beets and corresponding maximum wavelength $\lambda_{max.}$ of filtrate.

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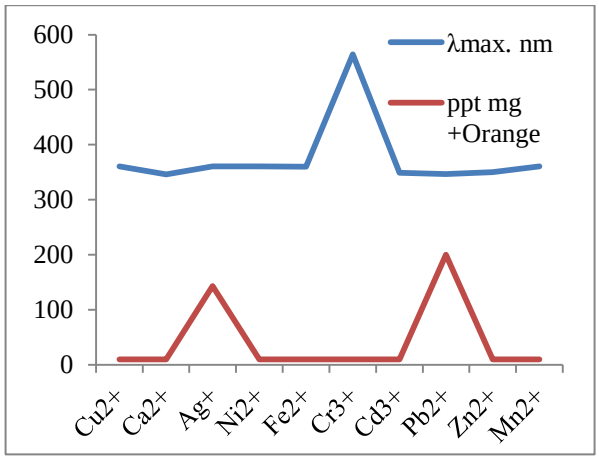


Figure 25. Shows the weight of metal ions precipitate in milligrams with Orange and corresponding maximum wavelength $\lambda_{max.}$ of filtrate.

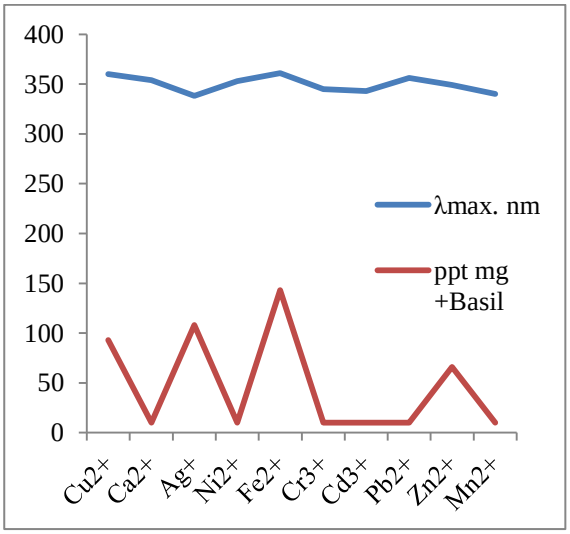


Figure 26. Shows the weight of metal ions precipitate in milligrams with Basil and corresponding maximum wavelength $\lambda_{max.}$ of filtrate.

5. CONCLUSIONS

There are Many organic compounds, from plants, could be used as ligands to chelate the heavy metal ions, this study used ten of natural product extraction as Cloves, Cinnamon, Sage, Mint, Maringa, Carrots, Cabbage, Beets, Orange and Basil to study the treatment of ten metal ions as Cu^{2+}, Ca^{2+}, Ag^{+}, Ni^{2+}, Fe^{2+}, Cr^{3+}, Cd^{3+}, Pb^{2+}, Zn^{2+} and Mn^{2+}. Then calculating the weight of precipitates and maximum wavelength of filtrate compare the maximum wavelength of metal ions and extractions, the constant $\lambda_{max.}$ values indicate that the quality of precipitation and used these extractions to treatment of waste water, water accompanied with oil and battery waste water.

6. ACKNOWLEDGEMENT

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