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Comparative Study of Lead Removal by Extracts of Spinach, Coffee, and Tea

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ABSTRACT

The use of tea and coffee for the removal of heavy metal from aqueous lead solutions has been reported. However, those studies were limited to expended dry biomass of coffee and tea and the lead concentration in those studies range from 10 - 100 ppm of aqueous lead solution. This study compared the effectiveness of aqueous extracts of instant coffee (IC), coffee ground (CG), coffee bean (CB), Lipton tea (Tea), and spinach puree (SP) in removing lead from 1300 PPM of aqueous lead solution. After 24 hr of agitation at room temperature followed by centrifugation, the lead concentration (in ppm) in the liquid from each reaction tube was analyzed using EPA Method 6010 (Inductively Coupled Plasma-Atomic Emission Spectrometry (ICP-AES)). The results suggest that the order of lead removal was Spinach (99%) > Instant coffee (95%) > Tea (91%), >CG (62%) > CB (59%). In comparing the brewed versus the boiled extracts, the results demonstrated that temperature of the aqueous extract affected the lead removal potential of coffee and tea in decreasing order: IC (95%:79%), >Tea (91%:88%) > CG (62%:53%) > CB (59%:53%).

Keywords: Heavy Metals; Tea; Coffee; Lead Remediation; Water Contamination; Phytoremediation

1. Introduction

Lead is a toxic contaminant with serious health implications to humans, animals, and plants. Lead has strong affinity for and complexes with many biomolecules and adversely affects the cardiovascular, nervous, gastrointestinal, reproductive, immune, renal, skeletal and muscular systems. In 1998, Johnson [1] reported that lead can cause cancer, birth defects, enzyme inhibition, and aberrations in DNA synthesis and mutation. Lead exposure to children has been associated with impaired cognitive skills [2], intellectual impairment [3], reduced mental IQ and quantitative skills [4], mild mental retardation and cardiovascular disease [5]. Chronic exposure of adults to lead can lead to neurological, reproductive system, and liver damage [6]; reduced cognitive function [7], and kidney disease [8]. Sadly, lead contamination from various sources is a global problem and represents a serious threat to soil, water, and air with a potential for propagation into the food chain. Thus, our global village is in need of innovative and cheap technology for the amelioration of lead contamination.

Presently, several technologies exist for the remediation of lead contaminated soil [9-11] and they include a) excavation and bury in landfills; b) capping; c) subsurface barriers; d) *in-situ* and *ex-situ* precipitation, solidification, stabilization; e) washing and f) biological treatment such as vermiremidiation [12], phytostabilization, phytodegradation or transformation, Phytochelation, and phytoextraction [13-16].

Although, few technologies such as electrodeposition and chemical precipitation exist for the remediation of lead contaminated water [17-19], they also have their limitations such as introduction of chemicals into the water and environment. Emerging research reports have shown that some substrates have the potential to remove lead from contaminated water [20-23].

However, the levels of contamination investigated were below 500 ppm and they used solid biosorbents such as waste solid tea leaves, coffee ground, or lignin. These adsorbents have the potential to increase the volume of solid waste in the water and environment. Thus, Agwaramgbo [24,25] reported the use of liquid extracts of edible plants for the exclusive removal of lead from contaminated water. It has been reported that plants including green tea, coffee, and green vegetables (spinach), contain polyphenols (tannins, catechins), gallic acid, and over one hundred other chemical compounds [26,27]. Tea and coffee also contain caffeine in addition to these other phytochemicals. These tannins or polyphenols are associated with the precipitation and removal of heavy metal from aqueous solutions [26-28]. The roasting process of coffee has a destructive effect on its constituent

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polyphenols (tannins) [28]. The research results presented here compare the lead removal abilities of the liquid extracts of spinach, brewed and boiled instant coffee, coffee ground, coffee bean, and Lipton tea.

2. Materials & Methods

2.1. Preparation of Lead Nitrate Solution 1300 ppm

Using an analytical balance, 1.3 g of lead Nitrate from Fisher Scientific (L6200) was dissolved in enough deionized water (added incrementally) to give 1000 ml of solution. Then a stirring bar was dropped into the volumetric flask and the mixture was stirred until all the lead was completely dissolved. The solution continued to stir at room temperature until it was used.

2.2. Preparation of the Spinach Extract

Spinach (100 g) bought from a local market; was washed with deionized water and patted dry with kimwipes. The spinach was pureed in a regular kitchen blender using 200 ml of de-ionized water and filtered using a white cheese cloth bought from a local Wal-Mart store. The filtrate was put into four-50 ml centrifuge tubes and centrifuged at 3000 rpm for 10 minutes using a Thermo Centra CL2 bench-top centrifuge. Using disposable pipettes, three-25 ml portions of each resulting supernatant was carefully transferred into three-50 ml centrifuge tubes, respectively. The tubes were capped, labeled, and placed in a refrigerator until use.

2.3. Preparation of Brewed Coffee and Tea Extracts

Community Dark Rose Instant Coffee (IC), Coffee Ground (CG), Coffee Bean (CB); and Lipton Tea (Tea) were bought from a local market. Each beverage (45 g) was respectively brewed with 250 ml of deionized water using a coffee brewing machine. The temperature of each brewed aqueous extract was 50˚C - 60˚C. Each brewed extract was placed in an Erlenmeyer flask, covered, labeled (IC-B, CG-B, CB-B, Tea-B), and placed in a refrigerator until use.

2.4. Preparation of Heated Coffee and Tea Extracts

Respectively, 45 g of Community Dark Rose Instant Coffee (IC), Coffee Ground (CG), Coffee Bean (CB); and Lipton Tea (Tea) were placed in 500 ml beakers. Deionized water (250 ml) was added to each beaker and its content. The mixture was heated to a boil at 90˚C - 91˚C for five minutes. Each mixture was filtered, and the filtrate was centrifuged at 3000 rpm for ten minutes. The supernatant was transferred into three 50-ml centrifuge tubes. Each set of triplicate centrifuge tubes for instant coffee, coffee ground, coffee bean, and tea were labeled (IC-H, CG-H, CB-H, and Tea-H), and placed in a refrigerator until use.

2.5. Preparation of Control Lead Solution

Into three 50 ml centrifuge tubes was added 50 ml of the 1300 ppm of lead nitrate solution above to serves as control. Each tube was labeled as Pb Ctr.

3. Reaction of Spinach and Brewed Coffee and Tea Extracts

3.1. Reaction of 1300 ppm of Lead Solution with the Spinach Extract

Into each set of triplicate centrifuge tubes containing 25 ml of the spinach extract was added 25 ml of the 1300 ppm of the lead nitrate solution prepared above. The three tubes and their contents along with triplicate tubes containing the 1300 ppm lead solution were vortexed, tightly secured on the rack of a heavy duty Eberbach 6000 shaker, and agitated for 48 hours at room temperature.

3.2. Reaction of 1300 ppm of Lead Solution with the Brewed Coffee Extracts (IC-B, CG-B, and CB-B)

Following the same procedure used in the reaction of spinach extract with lead solution above, 25 ml of the lead nitrate solution (1300 ppm) prepared above was added to each of the three sets of triplicate centrifuge tubes containing 25 ml of the brewed instant coffee, coffee ground, and coffee bean extracts, respectively. The nine tubes and their contents were vortexed and agitated for 48 hours at room temperature on a heavy duty Eberbach 6000 shaker.

3.3. Reaction of 1300 ppm of Lead Solution with the Brewed Lipton Tea Extract (Tea-B)

Following the same procedure used in the reaction of spinach extract with lead solution above, 25 ml of the lead nitrate solution (1300 ppm) prepared above was added to each of the three triplicate centrifuge tubes containing 25 ml of the brewed Lipton tea. The three tubes and their contents were vortexed, and agitated for 48 hours at room temperature on a heavy duty Eberbach 6000 shaker.

3.4. Reaction of 1300 ppm of Lead Solution with the Boiled Coffee Extracts (IC-H, CG-H, and CB-H)

Following the same procedure used in the reaction of brewed spinach extract with lead solution above, 25 ml of the lead nitrate solution (1300 ppm) prepared above was added to each of the three sets of the triplicate centrifuge tubes containing 25 ml of the boiled instant coffee (IC-H), coffee ground (CG-H), and coffee bean (CB-H)], respectively. The nine tubes and their contents were vortexed, and agitated for 48 hours at room temperature on a heavy duty Eberbach 6000 shaker.

3.5. Reaction of 1300 ppm of Lead Solution with the Boiled Lipton Tea Extract (Tea-H)

Following the same procedure used in the reaction of spinach extract with lead solution above, 25 ml of the lead nitrate solution (1300 ppm) prepared above was added to each of the three centrifuge tubes containing 25 ml of the boiled Lipton tea extract (Tea-H). The three tubes and their contents were vortexed, tightly secured on the rack of a heavy duty Eberbach 6000 shaker, and agitated for 48 hours at room temperature on a heavy duty Eberbach 6000 shaker.

3.6. Agitation of the Control Lead Solution

The three tubes containing the 1300 ppm lead solution prepared above were vortexed and agitated for 48 hours at room temperature on a heavy duty Eberbach 6000 shaker.

4. Sample Preparation and Analysis

4.1. Sample Preparation

After 48 hrs, the shaker was stopped and the tubes and their contents were centrifuged at 3000 rpm for ten minutes. The resulting clear supernatant in each tube was transferred into another labeled clean centrifuge tube. All the labeled centrifuge tubes with their liquid contents were sent to PACE Analytical Services, Inc for lead analysis.

4.2. Sample Analysis for Lead after Reaction

After the reaction period, the lead concentration (in ppm) in the liquid from each reaction tube was analyzed using EPA Method 6010 (Inductively Coupled Plasma-Atomic Emission Spectrometry (ICP-AES)).

4.3. Statistical Analysis

Use of descriptive statistics to interpret the data will be useful to determine if the extract isolation temperature is associated with the lead removing capacity of the extracts. Thus, Pearson correlation coefficient (r), percent variance (CV%) will be used to show if an association exists between the temperature of extract isolation and the lead removing capacity of the extract.

5. Results

Data on **Table 1** show residual lead in ppm in each reaction tube after contaminated water was treated with each extract for 48 hrs and the percent of lead removed relative to the control: Control (1280, 0%); IC-B (63, 95%, CG-B (488, 62%), CB-B (527, 59%), Tea-B (120, 91%, Spinach (7, 99%). **Figures 1(a)** and **(b)** show the amount of lead remaining in the reaction mixture after coffee, tea, and spinach extract treatment and the percent of lead removed, respectively.

6. Discussions

6.1. On Spinach Extract

The results on **Table 1** and **Figures 1(a)** and **(b)** showed that spinach removed over 99% of the lead from contaminated water and served as a reference for the brewed and boiled coffee and tea extracts.

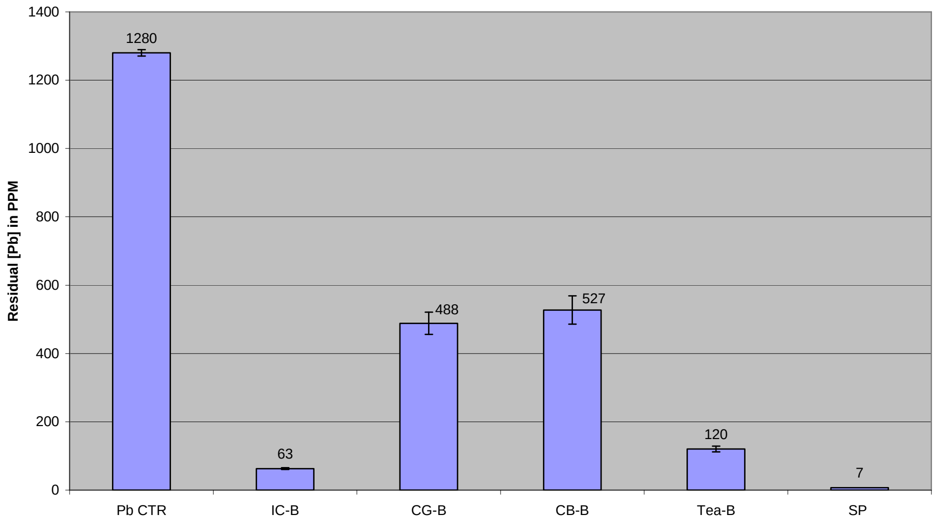
6.2. Comparison of Lead Removal by Brewed Coffee & Tea Extracts

Table 1 and **Figures 1(a)** and **(b)** clearly showed that spinach, instant coffee, and tea have unusually great ability to remove lead from contaminated water. However, ground coffee and coffee bean had a modest ability to remove lead from lead contaminated aqueous solution. The order of lead removal ability is: Spinach (99% > Instant Coffee (95%) > Tea (91%) > Coffee Ground (62%) > Coffee Bean (59%).

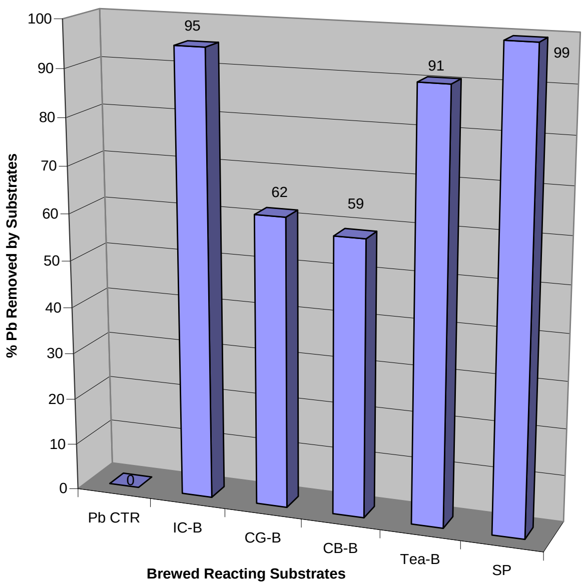
It is not surprising that instant coffee removed far more lead than ground coffee and coffee bean due to the fact that all the amount of instant coffee used solubilized in solution during brewing. Thus, instant coffee has more available lead-chelating or adsorbing agents in contact with the lead. However, in the case of ground coffee and coffee bean, only a fraction of the chelating chemical constituents of the solid biomass used solubilized in solution during brewing. Therefore, they had less amount

Table 1. Residual & removed lead in contaminated water after treatment with spinach and brewed coffee/tea extracts.

Sample	[Pb] in PPM	Std. Dev.	Std. Error	% lead removed
Control (Ctr)	1280	16.32	9.42	0
Instant Coffee (IC-B)	62.8	4.77	2.75	95
Coffee Ground (CG-B)	488	55.9	32.29	61.8
Coffee Bean (CB-B)	527	71.6	41.34	58.8
Tea (Tea-B)	120	14.64	8.45	90.6
Spinach (SP)	7.16	0.42	0.24	99.4



(a)



(b)

Figure 1. (a) Lead concentration remaining after contaminated water was treated with extracts of spinach and brewed coffee and tea; (b) Percent lead removed after contaminated water was treated with extracts of spinach and brewed coffee and tea.

of the lead chelating substances brewed extracts. It is worthy to note that even though equal amount of dry solid biomass of tea, ground coffee, and coffee bean was used, tea removed 40% - 50% more lead. It is possible to conclude that tea has more lead chelating agents per unit dry weight than coffee ground and coffee bean.

Figure 2 above showed that there is a strong association between plant type and their lead removal potential (R^2 of 0.7721 for brewed extracts and 0.8787 for boiled extracts).

6.3. Comparison of Lead Removal by Boiled Coffee and Tea Extracts

The data on **Table 2** and **Figures 3(a)** and **(b)** suggest that Tea extract removed more lead than instant coffee, while ground coffee and coffee beans removed equal amount of lead Thus, the order of lead removal is Tea (88%) > Instant Coffee (79%), Coffee Ground (53%) ≈ Coffee Bean (53%).

It is possible that instant coffee contains some volatile or semi-volatile lead chelating components that evaporated during heating.

6.4. Effect of Temperature on Lead Removal by Coffee and Tea: A Comparison of Brewed Versus Boiled Coffee & Tea Extracts

From **Table 3** and **Figures 4(a)** and **(b)**, it is apparent that lead removal by each extract is clearly affected by an

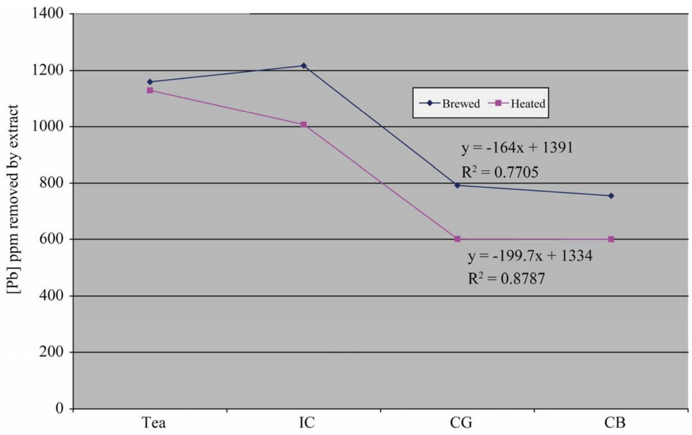
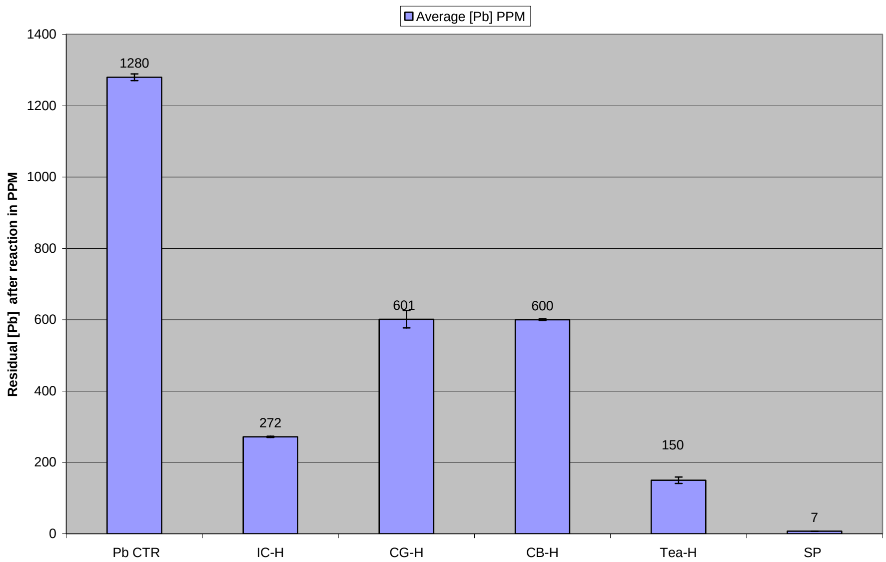
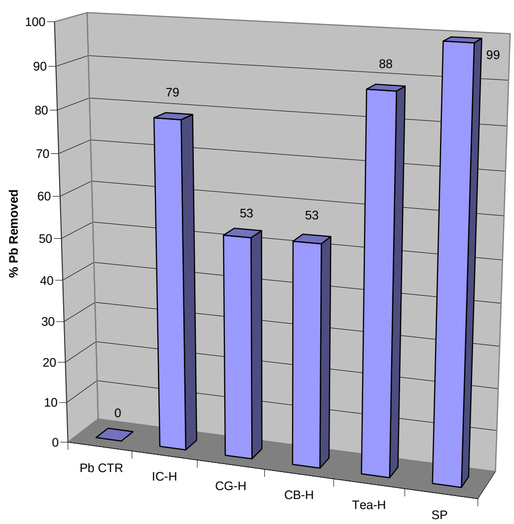


Figure 2. Line graph of data on Table 4: Comparison of lead removal of coffee and tea extracts isolated at 60˚C (brewed) & 90˚C (heated or boiled) R^2 (brewed) = 0.7705, & R^2 (heated) = 0.8787.

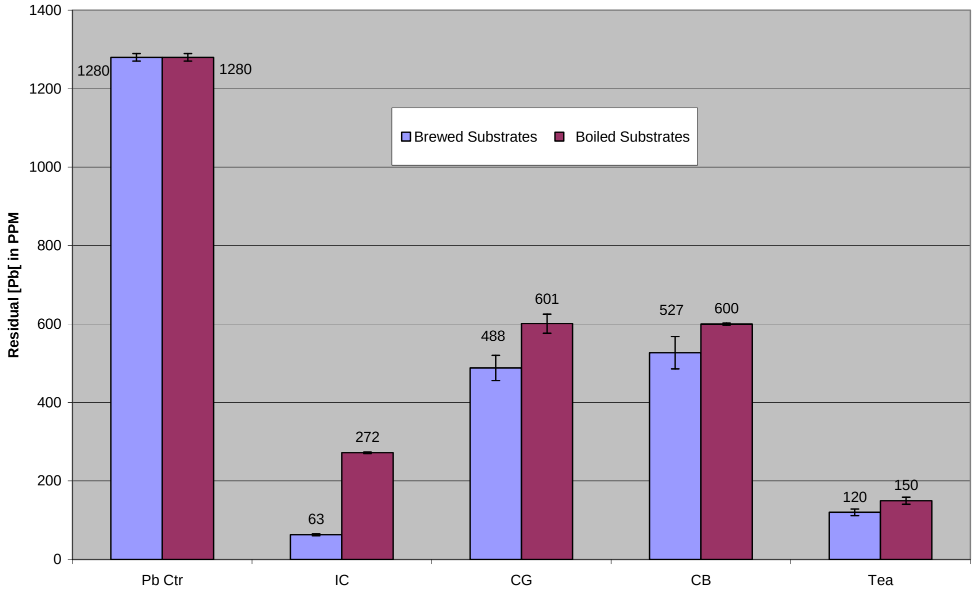


(a)

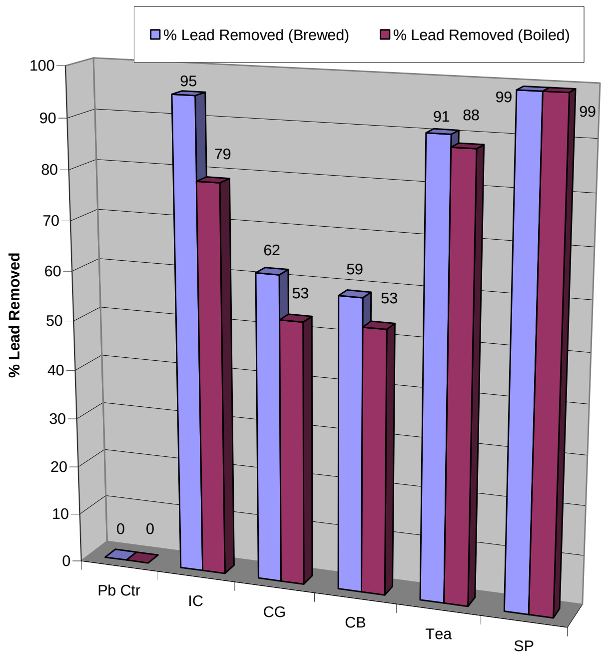


(b)

Figure 3. (a) Lead concentration remaining after contaminated water was treated with extracts of spinach and boiled coffee and tea; (b) Percent lead removed after contaminated water was treated with extracts of spinach and boiled coffee and tea.



(a)



(b)

Figure 4. (a) Lead concentration remaining after contaminated water was treated with extracts of brewed & boiled coffee and tea (IC-B, IC-H; CG-B, CG-H; CB-B, CB-H; Tea-B, Tea-H); (b) Percent lead removed after contaminated water was treated with brewed and boiled coffee and tea extracts (IC-B, IC-H; CG-B, CG-H; CB-B, CB-H; Tea-B, Tea-H).

increase in temperature of extraction. The data in **Table 3** suggest that heating the extracts from a normal brewing temperature of 60˚C to a boiling temperature of 90˚C effectively decreases the lead removal potential of all extracts reported in this paper.. Such a decrease was very steep in the case of instant coffee. The brewed extract from instant coffee removed 95% of the lead from the contaminated aqueous solution while the corresponding boiled extract (IC-H), removed 79% of the lead. Coffee Ground and Coffee Bean had a small but noticeable reduction lead removal as the temperature increased from 60˚C to 90˚C, a reduction of 9% and 6%, respectively (CG-B, 62% vs CG-H, 53%; CB-B, 59% vs CB-H, 53%). The data in **Table 4** has a Pearson correlation coefficient of 0.949 which suggests that there is a strong association between increase in temperature of extract isolation and decreased capacity of lead removal by the extract. Also, the data may demonstrate that tea contains more lead-

Table 2. Residual & removed lead in contaminated water after treatment with extracts of spinach and boiled coffee and tea.

Sample	[Pb] in PPM	Std. Dev.	Std. Error	% lead removed
Control (Ctr)	1280	16.32	9.42	0
Instant Coffee (IC-H)	272	3.39	1.96	79
Coffee Ground (CG-H)	601	42.1	24.3	53
Coffee Bean (CB-H)	600	4.5	2.6	53
Tea (Tea-H)	150	15.62	9.0	88

Table 3. Residual & removed lead in contaminated water after treatment with brewed and boiled coffee and tea extracts.

Sample	[Pb] in PPM	Std. Dev.	Std. Error	% lead removed
Control (Ctr)	1280	16.32	9.42	0
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Instant Coffee (IC-B)	62.8	4.77	2.75	95
Instant Coffee (IC-H)	272	3.39	1.96	79
Coffee Ground (CG-B)	488	55.9	32.29	61.8
Coffee Ground (CG-H)	601	42.1	24.3	53
Coffee Bean (CB-B)	527	71.6	41.34	58.8
Coffee Bean (CB-H)	600	4.5	2.6	53
Tea (Tea-B)	120	14.64	8.45	90.6
Tea (Tea-H)	150	15.62	9.0	88

Table 4. Comparison of lead removed from contaminated water by brewed and boiled coffee and tea extracts the Pearson correlation coefficient R = 0.949, R^2 = 0.9006, & CV = 90.06%.

Plant	X [Pb] removed by brewed extract	Y [Pb] removed by boiled extract	X^2	Y^2	XY
IC	1217	1008	1,481,089	1,016,064	1,226,736
Tea	1160	1130	1,345,600	1,276,900	1,310,800
CG	792	697	627,264	485,809	552,024
IC	753	680	567,009	462,400	512,040
Σ	3922	3515	4,020,962	3,241,173	3,601,600

Equation 1: Pearson Correlation Coefficient Equation

$$r = \frac{\sum XY - \left[\sum XY - \sum X \sum Y\right]/N}{\sqrt{\left\{\left[\sum X^2 - \left(\sum X\right)^2/N\right]\left[\sum Y^2 - \left(\sum Y\right)^2/N\right]\right\}}}$$

Substituting the values obtained from the **Table 4** above into the equation, gave Pearson's correlation coefficient of 0.949, with R^2 of 0.9001 and percent variance of 90.01%.

precipitating phytochemicals such as tannins and polyphenols than coffee. Furthermore, it has been reported that the roasting process of coffee has a destructive effect on its chemical constituents due to volatilization, decomposition, and denaturation as the temperature is increased [27,28]. Thus, the order of lead removal by extracts isolated at 60˚C is Instant Coffee (95%) > Tea (91%) > Coffee Ground (62%) > Coffee Bean (59%) compared to the lead removal by extracts isolated at 90˚C; Tea (88%) > Instant Coffee (79%) > Coffee Ground (53%) ≈ Coffee Bean (53%). It is worthy to note that instant coffee has most of its phytochemical contents extracted into the solution whereas not all the chemicals are extracted into the solution in the case of tea, coffee ground, and coffee bean.

7. Conclusions

Extracts of spinach, instant coffee and tea effectively removed over 90% of the lead in contaminated aqueous solutions. Among the brewed extracts investigated, instant coffee had the highest lead-removal ability followed by tea. However, upon increasing the extract isolation temperature from 60˚C to 90˚C, each ex tract showed a reduction in lead-removing potential, particularly instant coffee. This finding is in agreement with the Pearson correlation coefficient of 0.949, suggesting that increasing the temperature reduces the lead removing potential of the extract. These results demonstrate that instant coffee and tea extracts could provide a convenient technique for lead removal from contaminated waters. Furthermore, the results suggest 1) that the amount of biologically active components of these extracts that contribute to lead removal varies from one extract to another; 2) that elevated temperatures above 60˚C decrease the extracts' lead-removing potential. Such reduction may likely be in part, due to volatility, decomposition or denaturing of some of those chemical and biological functional groups in the extracts that play important role in lead complexation, precipitation, and adsorption [27,28].

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