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Removal of Toxic Metals from Water using Cabbage, Carrots and Lettuce as Biofilters

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Abstract

The existing technologies for removing toxic metals from the environment are very expensive and impractical for use in developing countries. In the present study we investigated the use of Lettuce (*Lactuca sativa*), Cabbage (*Brassica oleracea var. capitata*) and Carrots (*Daucus carota subsp. Sativus*) as biosorbents for removing Copper (Cu), Lead (Pb), and Zinc metal ions from polluted water via biofiltration. The filtrates were analysed via Atomic Absorption Spectroscopy (AAS) in order to quantitate any remaining, unremoved metals. All three biosorbents showed considerable abilities to remove the above metals, with about 95% removal efficiency in the case of zinc (Zn^{2+}). In order to understand the chemistry of the adsorption processes, the biosorbent materials, after biofiltration, were analysed via FT-IR spectroscopy, which revealed that all three biosorbents contained compounds with phenol, ether, alkanol, carbonyl and alkanoic acid groups that were involved in the formation of surface complexes during the adsorption of the metal ions. To investigate the effect of this biofiltration process on the overall quality of water, the physicochemical parameters (pH, conductivity, colour, turbidity, TDS and TSS) were also measured on river water samples that have been pre-treated with the biosorbents. There was general improvement in the water quality although the biofilters appeared to have introduced some mineral ions, which increased the conductivity and TDS of the water.

Keywords: Biofitration; Lead; Zinc; Copper; Remediation; Mining; Pollution

1. Introduction

Environmental pollution from chemicals remains one of the serious threats globally (Schwarzenbach et al. 2010), but the indiscriminate release of toxic heavy metals into soil and water bodies is much more alarming (Valko, Morris, and Cronin 2005) and requires utmost attention. Unlike some organic contaminants, heavy metals, which are inorganic, cannot be broken down to non-toxic forms and therefore have long-lasting effects not only on human health but the ecosystem at large. Even at very low concentrations heavy metals including lead (Pb), cadmium (Cd), mercury (Hg), chromium (Cr), zinc (Zn), silver (Ag), nickel (Ni) and arsenic (As) are well known to be cytotoxic and carcinogenic in nature (Gjorgieva Ackova 2018; Jia et al. 2017; Fashola et al. 2016; Tchounwou et al. 2012). Although heavy

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metals in general are naturally occurring, human activities, especially mining has caused exacerbating levels of these toxic metals (Delplace et al. 2020; Shi et al. 2018). Once released into the environment, toxic metals transfer and bioaccumulate in plants, tissues of animals, including freshwater fish, and eventually enter humans through the food chain, inhalation, or handling of contaminated samples (Delplace et al. 2020; Wuana and Okieimen 2011). The threats posed by these toxic heavy metals call for effective removal strategies in order to avert any unexpected health implications.

(Malik et al. 2016) have discussed a number of conventional techniques that can be used in removing heavy metals from the environment. Unfortunately, all such techniques have their limitations, especially with respect to high cost of operations, making them largely impractical for developing countries. The search for cheaper alternatives therefore continue to be sought by researchers worldwide. Biofiltration, a physico-chemical process, which involves the use of biological materials (plants and microbes) to remove heavy metals from aqueous solutions have been reported to be cost-effective and environmentally friendly solution compared to the more expensive conventional techniques (Majumder and Gupta 2010; Srivastava and Majumder 2008).

Plants, especially leafy vegetables grown on either contaminated soils or irrigated with polluted water tend to be a major source of toxic metal consumption by humans (Rai et al. 2019; Intawongse and R Dean 2006; Müller and Anke 1994). Researchers have exploited this metal-adsorption ability of such plants to use as potential biofilters for removing the same metals from the environment. Several studies suggest that leafy vegetables such as cabbage, lettuce and carrots accumulate large amounts of heavy metals even in the presence of high levels of plant nutrients (Al-Hiyaly 2014; Boamponsem et al. 2012; Mumba et al. 2008; Intawongse and Dean 2006; Zhou et al. 2005; Cobb et al. 2000). Vegetables have various mechanisms for taking up heavy metals from the environment, and the various parts (roots, stems, leaves, etc.) may accumulate different metals at different levels. For example, some plants accumulate more metals in the leaves than in the roots or stems, and vice versa (Othman 2001).

In the present study, the use of the edible parts of cabbage (i.e. head), lettuce (i.e. leaves) and carrot (i.e. root) as biosorbent materials for the removal of zinc, lead, and copper from artificially contaminated water as well as river water samples was investigated. The study was carried out in Sunyani in the Bono region of Ghana, and spanned the period from September 2018 to April 2019.

2. Materials and Methods

2.1 Reagents and Solutions

All reagents used were analytical grade. Lead oxide (PbO), Zinc oxide (ZnO) and Copper sulphate ($CuSO_4$) were purchased from BDH Laboratory, England, and used as is. Solutions of the heavy metal ions, Lead (Pb^{2+}), Zinc (Zn^{2+}) and Copper (Cu^{2+}) were prepared through appropriate dilutions from a 1000 ppm stock solution of the respective compounds listed above. The solutions were prepared with deionized water and supplemented with 10% (v/v) nitric acid (HNO_3) to facilitate complete dissolution. Where necessary, 0.1 M solutions of nitric acid (HNO_3) and sodium hydroxide (NaOH) were used to adjust the pH as appropriate.

2.2 Preparation of the Adsorbent/ Biofilter

The vegetables were obtained from a selected farm at Abesim Dadaa, near Abesim in the Sunyani Municipality of the Bono Region of Ghana. The fresh vegetables, carrots (roots), lettuce (leaves), and cabbage (head) were thoroughly washed under running tap water and further rinsed with distilled water in the laboratory. The vegetables were then chopped into smaller pieces and air-dried for about 30 min to allow any moisture to dry out. Afterwards, they were placed in an oven and further dried at 70 ^{o}C for 12 h. The dried vegetables were then ground to fine powder using a mechanical blender. These powdered samples were transferred into zip-lock bags and stored airtight at 4 ^{o}C until ready for use.

2.3 Biosorption Experiments

The removal of Pb^{2+}, $Zn^{2+}$and Cu^{2+} ions from the artificially contaminated water were investigated through adsorption process involving incubation of the metal ion solutions with the biosorbents (powdered vegetable samples) followed by filtration and quantitation. Specifically, 100 mg of the finely grinded vegetable was added to 20 ml of the metal solution in a pre-treated flask. It was then incubated at 40°C for about 1 h with gentle shaking at 300 rpm. The suspension was then filtered using filter paper (Whatman Nº1). Residual metal ion (Pb^{2+}, Zn^{2+} or Cu^{2+}) remaining in the filtrate was then quantified via flame atomic absorption spectrophotometry (FAAS) using the Analytik Jena AAS Model AA400p instrument, Germany. The ability of the biosorbents to remove the specific metals were determined as percent removal, and calculated as follows:

$$Removal\ (\%) = \frac{Concentration\ \ before\ filtration\ (ppm) - Concentration\ \ after\ filtration\ (ppm)}{Concentration\ \ before\ filtration\ (ppm)} \times 100$$

2.4 IR Characterization

The biomass residues after the biofiltration process were air-dried and analyzed for their functional group properties via Fourier-Transform Infra red spectroscopy (FT-IR). Samples of the dried biomass powder prior to the biofiltration were also analyzed with FT-IR in order to compare and evaluate any changes in functional group properties before and after the biofiltration.

2.5 Application of the biofiltration process on raw water samples.

The ability of the biosorbents to improve the physicochemical quality of water was also tested on water samples collected from the Kwabrafo River in Obuasi, a well-known mining town in Ghana. The Kwabrafo River is defined by the presence of mining activities such as tailing dams both active and inactive, gold-ore crushing and processing facilities, and purportedly contaminated with solid waste from the Obuasi township (CHRAJ 2008). The stream has a turbid brownish colour with unpleasant smell, but still used by the inhabitants for various domestic purposes. The biofiltration process described above was carried out on these water samples for all three vegetables. The raw water sample was analyzed via FAAS, as described above, to determine the presence of the metals (Pb^{2+}, Zn^{2+} or Cu^{2+}). The various filtrates from the biofiltration process were also analyzed for the physicochemical parameters pH, electrical conductivity, turbidity, colour, total dissolved solids and total suspended solids. The electrical conductivity and Total Dissolved Solids (TDS) were measured using E-1 portable EC / TDS meter (Newly Digital, China). pH of the sample was measured with pH meter (pH901, Bante Instruments, China). The remaining parameters were analyzed using standard protocols.

3. Results and Discussion

3.1 Removal efficiencies

The FAAS was used to quantitate the actual concentrations of the metals present in the artificially contaminated water and to also determine the concentrations of these metals remaining after the biofiltration process.

Figure 1 provides a comparison of the removal efficiencies of the three biosorbents - carrots, cabbage, and lettuce towards the three heavy metals zinc, lead, and copper.

All three biosorbents showed generally the same trend for uptake preference towards the three metals, with all biosorbents showing highest removal efficiency towards zinc, followed by copper, and then lead. About 95% (cabbage, 95.24%; carrot, 95.26%; lettuce, 95.22%) of the initial 100 ppm zinc was removed after the biofiltration process. The uptake of copper by the three biosorbents were also very similar, with carrots removing 67.74% of the initial 100 ppm; followed by 67.71% and 67.00% respectively for cabbage and lettuce. The only difference in removal efficiencies among the three biosorbents was observed for the uptake of lead. Carrot showed the highest preference for lead, with 34.76% of the initial 100 ppm removed after biofiltration. Cabbage showed an uptake efficiency of 29.81%, with the least preference for lead observed in lettuce, which showed removal efficiency of only 11.14%. Generally, all three biosorbents showed relatively poor preference towards the uptake of lead. The similarities in the percent removals may, however, suggest very similar chemical constituents present in all of three vegetables. Zinc is quite mobile and readily taken up by plants; whereas copper and lead are known to be strongly adsorbed to soil and other organic matter, making them less available for adsorption by plants (Usman 2008).

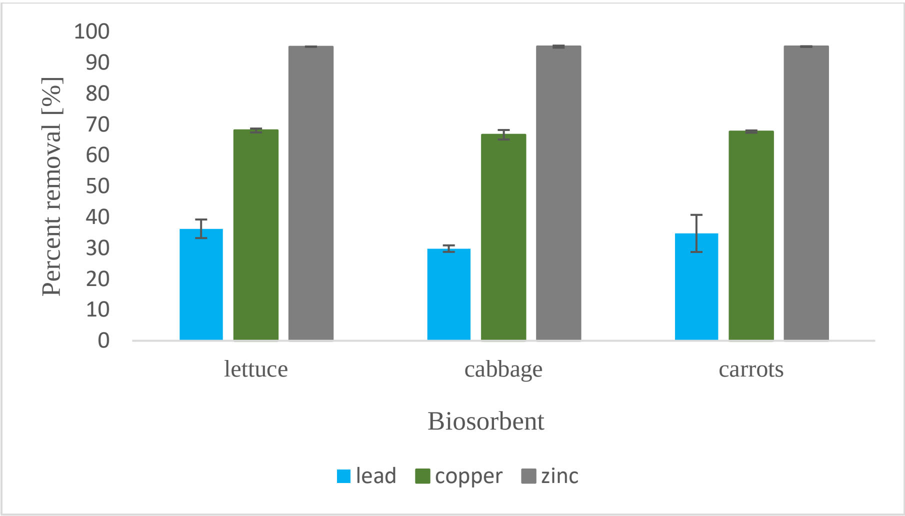

Fig. 1: Performance of the biosorbents cabbage, lettuce, and carrot towards the removal of zinc, lead, and copper from water. Performance was evaluated as percent removal based on the percent of metal ions removed by the specific biosorbent from the initially 100 ppm contaminated water

Several studies investigating the ability of vegetables to either uptake heavy metals from contaminated soil (bioremediation) or remove the metals from contaminated water (adsorption/ biofiltration) have been reported (Al-Hiyaly 2014; Chang et al. 2014; Boamponsem, Kumi, and Debrah 2012; Khan et al. 2008). However, to the best of our knowledge, no single paper has yet reported a thorough investigation on the chemistry and ability of cabbage, lettuce, and carrots to not only remove lead, zinc and copper from contaminated water but to also improve on the physicochemical quality parameters considered in this study. Heavy metal contamination of water is a serious global issue that adversely affects biota and poses long-term health risks. Since, the cost of operation encountered in the conventional methods of removing toxic metals from water poses a significant challenge, the use of low-cost, easily accessible plant materials have attracted wide attention, and researchers continue to explore effective biomaterials for removing these metals without compromising on other water-quality aspects (Malik, Jain, and Yadav 2016).

As a precaution to ensure that no pre-existing metals compromise with the biofiltration process, the powdered biosorbent materials were taken trough acid digestion to ensure that any trace amounts of metals were completely "destroyed" prior to the biofiltration experiment. The edible parts of the vegetables (i.e. root for carrot, leaf for lettuce, and head for cabbage) were purposively selected in order to help evaluate the extent to which these edible plants can accumulate the heavy metals, and its subsequent implications on consumers.

The FAAS results showed that all three vegetables have abilities to adsorb and therefore remove the three metals from polluted water. The high removal efficiency (95%) recorded for the zinc is consistent with literature (Moreira et al. 2015; Zwain, Vakili, and Dahlan 2014). The fact that all three biosorbents behaved similarly with respect to the removal efficiencies of the three metals may indicate similar chemistry of these adsorbents.

3.2 Characterization of the surface functional groups via IR spectroscopy

Just as observed in the results of the FAAS analysis, the FT-IR spectra resulting from the three biofilters are also identical. Figure 2 shows the IR spectra of the biosorbent, cabbage before and after the biofiltration.

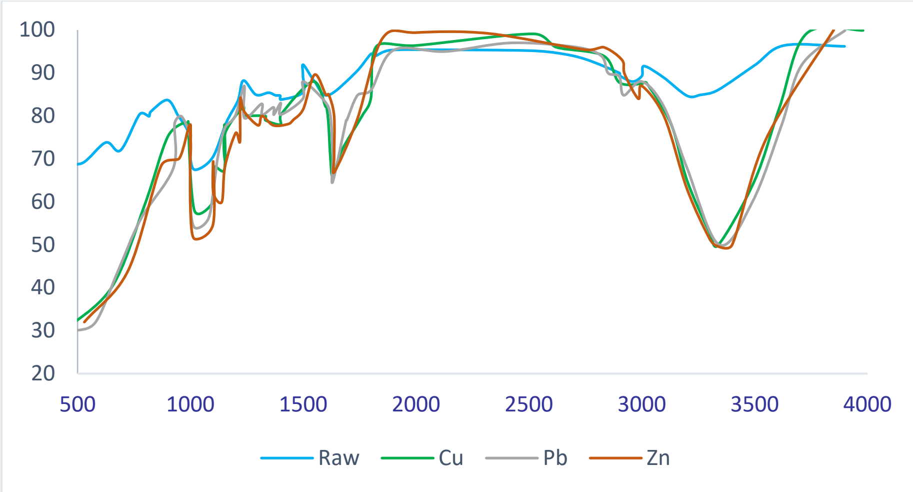

Fig. 2: Comparison of FT-IR spectrum of cabbage biosorbent before biofiltration and after biofiltration of lead, copper and zinc metal ions

Both spectra look identical to the counterparts for carrots and lettuce (not shown here). A broad band around 3200 cm^{-1} is observed in the spectra of all three biosorbents before the biofiltration, which shows O-H stretching vibrations suggesting the presence of phenolic groups, carboxylic acids, or alcohols. After biofiltration, this broad peak shifted to a higher frequency (3300 cm^{-1}) for all three biosorbents. These bands were also found to be narrow with a reduced transmittance ranging between 52 and 55% compared to the about 80-85% in the case of the biosorbents prior to the biofiltration process. These significant changes are clear indications that such functional groups were involved in the removal of the metals, and therefore compounds containing phenolic, carboxyl or alcohol groups are likely present in all three biosorbents. The peak around 2900 cm^{-1}, which also appeared in the spectra of all three biosorbents, is due to aliphatic C–H stretching vibrations of alkyl groups present. There was virtually no change in the frequency and transmittance after the biofiltration for all three biofilters. This suggests that the alkyl groups played no role in the removal process. The peak around 1600 cm^{-1} appearing in all the spectra of the biosorbents before the biofiltration is due to the stretching vibration of the C=O or aromatic C=C. After biofiltration, the peak appeared at a higher frequency around 1630 cm^{-1}, strongly suggesting that the C=O or aromatic C=C is involved in the removal of copper, zinc and lead ions. The presence of a sharp peak around 1015 to 1035 cm^{-1} in all the spectra of the biosorbents before biofiltration is due to C-O-C and C-O stretching vibration of ether and alcohols or phenols (He et al. 2017). After the biofiltration, the sharpness disappeared and the peak now appeared at a lower frequency.

Despite the identical spectra for all three biosorbents, the IR spectra for lettuce and carrot, before biofiltration, showed distinct peak at 1730 and 1742 cm^{-1} respectively, while it was completely absent for the cabbage biosorbent. In both cases, this peak is due to C=O stretching vibrations. Whereas this peak remained after the removal of copper with the lettuce biosorbent, it disappeared when the lettuce was used to remove lead and zinc. This may therefore suggest a slightly different chemistry or additional mode of interaction of the lettuce biosorbent with lead and zinc but not for copper. Such mode of interaction may also be available to the carrot biosorbent, although in this case, such particular group interacted with all three metals, as evident from the disappearance of the 1742 cm^{-1} peak after biofiltration.

The observation that all three biosorbents behaved similarly with respect to the removal efficiencies of the three metals, as confirmed with the FAAS analyses, is consolidated by the results of the FT-IR characterizations. The IR spectra of all three biosorbents before and after biofiltration appear similar. The shift in the O-H stretching vibrations to a higher frequency observed in the IR spectra of all three biosorbents is in line with what has been reported for similar biofiltration processes (Li et al. 2018). The aliphatic C–H stretching vibrations, which appeared around 2900 cm^{-1}, in both the before and after biofiltration residues, without any changes in frequency or transmittance is expected, as such alkyl groups do not show any significant involvement with metal ion interactions (Rostamian et al. 2015). The change in frequency or absence of a peak suggests that π electrons were likely involved in complexing with the metals during adsorption by the biofilters. It can therefore be concluded that the functional groups phenol, ether, alkanol, carbonyl and alkanoic acid groups were likely present and involved in the formation of surface complexes during the adsorption of Zn^{2+}, Cu^{2+} and Pb^{2+} by the biofilters carrot, cabbage and lettuce.

3.3 Physicochemical analysis

Considering that the biofilters were able to remove the various metals tested, it was important to evaluate any physicochemical changes that the adsorbent may have imparted on the quality of the water. In this regard, real water samples from the Kwabrafo River at Obuasi in the Ashanti Region of Ghana were taken and the biofiltration process was respectively applied to these samples. The physicochemical parameters (pH, conductivity, turbidity, TDS, TSS, and colour) were tested on the water samples before and after biofiltration. Table 1 shows results of the physicochemical parameters measured on the water samples before and after biofiltration.

Table 1: Physicochemical properties of water samples treated with and without biosorbents

Parameter	**Raw water**	**Water treated with biosorbent**			**Control***	**WHO Guideline****
		Cabbage	Carrot	Lettuce		
Ph	7.415	6.608	6.953	6.917	7.749	6.5-8.5
Conductivity(µs/cm)	513	1065	1075	1285	555	200 - 800
Turbidity (NTU)	12	4.3	6.5	4.8	10	< 5
Colour	18	12	14	13	17	<15
TDS (ppm)	256	532	545	598	177	< 1000
TSS (mg/l)	86	10	8	6	47	Not mentioned

*control = raw water filtered with Whatman No. 1 filter paper

**2017, WHO Guideline for drinking-water quality

The parameters selected are among the WHO's operational monitoring parameters, which can provide timely indication about the improvement or deterioration of river-water quality (WHO 2017). Apparently, the biosorbent-treated water show quite an improvement in the major parameters measured compared to the raw, untreated water. The increases in conductivity as well as total dissolved solids (TDS) in the treated water samples compared to the raw water and the control are not surprising. Cabbage, carrot, and lettuce being edible vegetables contain important nutrients such as potassium (K^{+}), Calcium (Ca^{2+}), and magnesium (Mg^{2+}) ions in addition to antioxidants and other water-soluble vitamins (Warman and Havard 1997). All these ions can contribute to an increased ability of the water to conduct electrical current, as evidenced in the conductivity values recorded. Although the recorded values are above the limit recommended by the WHO guidelines for drinking water, it must be emphasized that a higher conductivity does not necessarily indicate poor "palatability" of water but rather dependent on the nature of the dissolved ions. For toxic ions dissolved in water, a higher conductivity recorded is a good warning sign to avoid drinking such water. The control included in this physicochemical measurements was based on the use of filter paper (Whatman N°1), with no biosorbent, for filtering the water. The fact that the results obtained for this control are not much different from those of the raw water are clear indications of enhanced improvement of the water quality in the presence of the biofilter. Thus, the biofilters were not only good at removing the heavy metals but were useful in enhancing the "palatability" of the water. However, the authors would like to caution that the results obtained here do not guarantee safe drinking water treated with these biosorbents, since additional research involving the toxicity and other healthrelated information may be required in order to draw a firm conclusion. The authors are of the view though that the results discussed in this work will provide significant contribution for future research aimed at developing effective, low-cost biosorbent materials for treating wastewater for irrigation purposes.

4. Conclusion

The ability of dried, finely powdered samples of the edible parts of cabbage (head), lettuce (leaves), and carrot (roots) to be used as biosorbents for the removal of zinc, copper and lead from the aqueous environment were investigated using water samples artificially contaminated with 100 ppm of the respective metals. As much as the biosorbents were able to adsorb the metals from solution, the biosorbents were also applicable for use on river water samples to help improve the overall quality. The use of the FT-IR to characterize the surface of the biosorbents before and after biofiltration was an important aspect of this study. Understanding the chemistry involved in the adsorption of these metals can lead to the development of novel adsorbents that are not only cost-effective but sensitive and selective towards various toxic metals. This work therefore represents a significant contribution to the search for efficient and cost-effective adsorbents in wastewater treatment technologies.

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