

Effects of Solution Matrix on Moringa oleifera Seeds and Banana Peel in Eliminating Heavy Metals, Fluoride and Turbidity from Synthetic Groundwater Samples

Nur Adila Ab Aziz^{1*}, Niranjali Jayasuriya², Linhua Fan², Mohd Azuan Zakaria¹, Faizal Pakir¹, Zawawi Daud¹, Adeleke Abdulrahman Oyekanmi³, Norli Ismail³

¹Faculty of Civil and Environmental Engineering, Universiti Tun Hussein Onn Malaysia, 86400 Batu Pahat, Johor, Analaysia

²School of Engineering, RMIT University, VIC 3000, Australia.

³School of Industrial Technology, Universiti Sains Malaysia, Pulau Pinang 11800, Malaysia.

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Abstract: Plant-based biomass has become an environmental-friendly water purification agent in replacing conventional chemicals. In the previous study, *Moringa oleifera* (MO) seeds and banana peel (BP) have been selected based on their moderate to high effectiveness in removing lead, cadmium, nickel, arsenic, turbidity, and fluoride from synthetic groundwater samples. This study was aimed to investigate further the effects of solution matrix on the biomass effectiveness. Batch experiments were conducted by using coagulation technique and the initial pH of the solutions was controlled to be at pH 7. The results demonstrate that the removal rates for most of the pollutants in multi-contaminant solution were higher compared to the single-contaminant solution. The reason could be due to electrostatic or mutual interactions between contaminants present in the solution thus improved the removal rates of those contaminants. In terms of performance, combination of MO and BP either dosed in mixing (MO+BP) or sequential manner (i.e., MO→BP or BP→MO) are seen can improve the treatment performance by single biomass for most of the target contaminants. The findings are significantly important to understand the effects and removal behavior of the biomass in different solution matrix.

Keywords: *Moringa oleifera*, banana peel, heavy metals, fluoride, turbidity, solution matrix, single-contaminant, multi-contaminant

1. Introduction

Plant-sourced materials have become an economical option in substituting expensive chemicals that conventionally employed in water treatment process. Natural water purification agents could be afforded by rural communities in some affected regions in South Asia countries as they are freely available [1].

In rural areas, people are relying on groundwater as their primary source of water that used for hygiene, daily activities and drinking purposes. Groundwater is often regarded to possess acceptable quality compared to surface water which can easily get contaminated by surrounding pollution contributing from uncontrolled storms and runoff events. However, according to Ul-Haq et al. [2], the sources are now vulnerable to the various harmful chemicals that may introduce by the discharge of untreated effluents from industrial activities and leach from pipeline materials into the groundwater.

For instance, the maximum concentrations of lead (43.2 ppb), nickel (25.9 ppb) and cadmium (3.9 ppb) were reported in Pakistan [3-4] while the mean

concentration level of arsenic was recorded to be 40 ppb in Sri Lanka [5]. These concentrations are exceeded the WHO drinking water quality standards of 10 ppb, 20 ppb, 3 ppb and 10 ppb for lead, nickel, cadmium, and arsenic, respectively. On the other hand, the maximum concentrations of fluoride were recorded significantly high in some rural parts of Sri Lanka, India, and Pakistan with 9.2 ppm, 6.6 ppm, and 4.9 ppm, respectively [5-7]. These concentrations exceed the WHO standard of 1.5 ppm for fluoride. There are numerous detrimental impacts caused by the prolonged ingestion of these harmful chemicals including cancer, skin lesions, cardiovascular disease, neuro-developmental and dental fluorosis [8-12]. Therefore, there is a need to eliminate these harmful chemicals before cooking and drinking to avoid the adverse impacts on health.

To meet the economic feasibility of the idea of introducing a decentralized drinking water treatment that could be employed by less skilled personnel in remote areas is via coagulation technique with biomass taking place as natural water purification agents substituting alum and ferric as conventional chemicals. There are substantial numbers of biomass have been explored for water treatment via various techniques including *Moringa oleifera*, *Cicer arietinum*, *Musa cavendish* (banana peel), *Lentinus edodes*, *Eichhornia crassipes*, and blue pine, walnut [13–18]. *Moringa oleifera* seeds and banana peel were proved to be moderate to highly effective in reducing our target contaminants such as heavy metals (lead, nickel, cadmium, and arsenic), fluoride and turbidity from synthetic and actual groundwater samples [19-20]. The lacking information from those works was the study on the effects of solution matrix (i.e., single- or multi-contaminant solution) on the selected biomass in removing target contaminants from the synthetic groundwater samples.

To the best of our knowledge, there is little evidence in the literature reporting on the effects of solution matrix on the effectiveness of biomass in removing heavy metals, turbidity, and fluoride. The outcomes are significantly essential to improve understanding of the effects and removal behavior of the biomass in different solution matrix. The information also might be useful for future treatment improvements.

2. Materials

2.1 Preparation of biomass

The seeds of *Moringa oleifera* (MO) seeds were supplied by Pacific Blue Consulting Pty. Ltd. (The Moringa Shop, Australia). The good quality of seeds was chosen, and the debris was removed from the seeds. The seeds were dehusked as recommended by [21]. The MO seeds were then washed with deionized water and dried in an oven at 100°C for an hour. The dried seeds were pulverized to powder form and passed through stainless steel mesh with an opening size of 300 μ m to standardize the particle size [22].

On the other hand, the peels of banana (BP) were gently removed from the fruits. The cleaned peels were then dried in an oven for an hour at 100°C. Subsequently, the peels that have been dried were cut into small pieces, pulverized and sifted through a stainless steel mesh with an opening size of 300 μ m to obtain the same particle size [23]. Stock solutions of MO and BP were promptly prepared prior the experiments by dosing one gram of each biomass, separately to the 1-L deionized water which made up a stock solution of concentration 1000 ppm, respectively. For the subsequent experiments, the desired dose of biomass was directly taken from those prepared biomass stock solutions.

2.2 Preparation of synthetic groundwater samples

The chemical stock solutions such as lead carbonate (PbNO₃), nickel carbonate (NiCO₃), cadmium nitrate tetrahydrate (Cd(NO₃)₂.4H₂O), arsenic trioxide (AsO₃), sodium fluoride (NaF) and kaolin powder (Al₂Si₂O₅(OH)₄) were obtained from Sigma Aldrich (Australia). Prior to single- and multi-contaminant samples preparation, stock solution for every chemical was prepared depending on the desired concentration

(determined based on the previous experimental results as presented in [1] & [2]. The concentrations of each target contaminant are presented in Table 1.

Table 1: Initial characteristics of single- and multicontaminant synthetic groundwater samples

containmant synthetic ground water samples					
Target	Initial	WHO drinking water			
contaminant	concentration	quality standard			
Lead	20 ppb	10 ppb			
Nickel	30 ppb	20 ppb			
Cadmium	5 ppb	3 ppb			
Arsenic	20 ppb	10 ppb			
Turbidity	15 NTU	5 NTU			
Fluoride	3 ppm	1.5 ppm			
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Note:

ppm = mg/L, $ppb = \mu g/L$, single-contaminant solutions (individually prepared), multi-contaminant solutions (all target contaminants were mixed together)

2.3 Batch experiments

The experimental investigations were conducted using a standard jar tester (Lovibond, Phipps & Bird). The pH of the sample was controlled to be 7.00 ± 0.02 using either solution of 0.1 M of sulfuric acid or 0.1 M of sodium hydroxide.

Two types of solution matrix were tested throughout the study namely single- and multi-contaminant synthetic groundwater samples. For single-contaminant solutions, the target contaminant (desired concentration as tabulated in Table 1) was added to the deionized water). Likewise, the same method was employed on multi-contaminant solutions to prepare the intended concentrations of target contaminants.

For the treatment process, three methods of biomass dosing were attempted. Biomass was dosed individually for single dosing method while in the mixing method, the biomass was added to the sample concurrently. For the sequential biomass dosing method, the addition of second biomass was subjected after the first sequence of treatment using the first biomass was done. The samples were subjected to rapid mixing (150 rpm for 3 minutes) and slow mixing (30 rpm for 30 minutes) before the agitated suspensions were allowed to settle for 30 minutes. The supernatant layer was taken to filtration with Advantec fiberglass membrane filter of pore size of 0.45 µm to determine the residual concentrations of the target contaminants. The optimum biomass doses used for this study were 200 ppm for individual dosing and 400 ppm (200 ppm: 200 ppm) for mixed or sequential dosing, respectively, as determined in our previous studies [18] & [12]. All experiments were carried out in triplicate (N=3), and the reported results are based on the mean values.

2.1 Analytical method

Concentrations of heavy metals, fluoride, turbidity, and pH were measured using Agilent 7700 series Inductively Coupled Plasma Mass Spectrometry (ICP-MS), HQ14d portable with ISEF121 Fluoride Ion Selective Electrode, HACH turbidity meter 2100AN and Seven Compact Mettler Toledo pH meter, respectively. Materials safety and data sheets (MSDS) were clearly understood prior to handling the chemicals and experiments. All analyses were carried out according to [17].

3. Results and Discussion

The effects of solution matrix on the removal rates of target contaminants (i.e., lead, nickel, cadmium, arsenic, turbidity and fluoride) were investigated in this study. The finding will be useful to evaluate the treatment performance of the selected biomass which mimicking the actual samples. In the actual samples, one contaminant may not present alone. There might be presence of other elements (i.e., physical, chemical or biological contaminants).

3.1 Heavy metal (cation) removal

In general, there are variations in terms of removal rates for all target contaminants across all attempted biomass dosing methods. Therefore, it is recommended to further study the removal behavior of target contaminants under single- and multi-contaminant conditions to ensure that the selected biomass was capable of eliminating target contaminants under actual environment [18].

In Fig. 1 to 3 (on the x-axis), single biomass dosing method represents by the abbreviation MO (Moringa seed) and BP (banana peel), respectively, while MO+BP represents the combined dose of both MO and BP in the ratio of 1:1. For instance, 200 ppm of MO was dosed concurrently with 200 ppm of BP, hence the total dose for MO+BP is 400 ppm. The sequential dosing method (i.e., MO \rightarrow BP and BP \rightarrow MO) means that the first biomass was dosed in the first treatment sequence, followed by the dosing of the second biomass to the withdrawn supernatant layer of the treated sample by the first biomass for further treatment.

Fig. 1a exhibits that lead was effectively removed by MO alone with 86% (multi-contaminant) and 81% (single-contaminant). It is worthy of note that, most of lead residual concentrations are complied with the WHO standard of 10 ppb (for lead) except for BP (single-element), thus suggests that MO alone is effective for lead removal.

For nickel (Fig. 1b), the highest removals (77%: single-contaminant and 67%: multi-contaminant) were achieved by the treatment with MO+BP. The residual concentrations of nickel were all met the WHO standard of nickel (20 ppb) after the treatment with both biomass and all attempted biomass dosing methods.

For cadmium (Fig. 1c), MO+BP showed the highest removal rate (77%) under single-contaminant condition. However, in multi-contaminant condition, the removal rates were lower to that of produced by MO+BP (67%) under single-contaminant condition, suggesting that the interference of cadmium with other elements in the system thus reduced the removal efficiency. All residual concentrations of cadmium were met the WHO standard of 3 ppb (for cadmium), thus proved the effectiveness of MO and BP for cadmium removal.

The removal rates for lead (Fig. 1a) and arsenic (Fig. 1d) were higher in multi-contaminant for all tested

biomass dosing methods. However, for nickel and cadmium, treatment with MO+BP and MO \rightarrow BP demonstrate higher removal for nickel and cadmium under single-contaminant condition. This finding suggests that the removal of lead and arsenic could be assisted by the mutual interaction between the electro charges of cations (heavy metals), anion (fluoride) and suspended matter (kaolin powder that represented as turbidity) under multi-contaminant condition.



Fig. 1 Residual concentrations and removal rates of (a) lead, (b) nickel, (c) cadmium and (d) arsenic using MO and BP dosed in different dosing methods

For instance, the removal of cadmium by MO+BP was higher under single-contaminant (Fig. 1c) could be due to the lower electronegativity value of 1.46 (for cadmium) compared to the other heavy metals which have higher electronegativity values as tabulated in Table 2 [26]. Electronegativity value indicates the ability of ion to attract bonding electrons towards it. This could be a hindering property possess by nickel and cadmium thus making them better removed by the tested biomass under

single-contaminant condition rather than in the competitive condition (multi-contaminant). This finding is in agreement with a finding reported by Vijayaraghavan et al., [27] which confirmed that the electronegativity is one of the factors that determine the preference of cations (i.e., lead, copper, manganese, zinc) attachment onto biomass (*Sargassum*). Futhermore, Zhu at al., [28] also found that this condition could be attributed by antagonistic effects between multi elements in the system.

Table 2: Physicochemical properties of heavy metal

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Properties	Lead	Nickel	Cadmium	Arsenic
Atomic weight	207.2	58.693	112.411	74.922
Electronegativity	1.55	1.75	1.46	2.20
Charge	2^{+}	2^+	2^{+}	3+

For arsenic, different MO and BP dosing methods produced almost similar removal efficiencies for arsenic which were around 40 to 42% under multi-contaminant condition (Fig. 1d). However, the increment of arsenic removals was seen when MO and BP were dosed in mixing and sequential manners. This finding reveals the actual performance of MO and BP when arsenic present in the system without interference with other elements. It also worthy of note that none of the arsenic residual concentrations were complied with the WHO standard of 10 ppb (for arsenic), indicating the MO and BP are ineffective for arsenic removal.

3.2 Fluoride (anion) removal

Fig. 2 demonstrates the removals and the residual concentrations of fluoride. The highest removal of fluoride was achieved by the treatment with the combination of MO and BP (MO+BP) with 42% (singlecontaminant) and 23% (multi-contaminant) removals. Also, it can be seen that the removal of fluoride were slightly improved when both biomass (MO and BP) were dosed either in mixing or sequential manner compared to the MO or BP alone. The residual concentrations of fluoride did not comply with the WHO standard of fluoride (1.5 ppm) for all attempted biomass and dosing methods. The reason could be the heavy metals which carry the opposite charges to that of fluoride which is anion (negatively charged ion), thus hindering the mutual electrostatic interactions which might assist the removal of fluoride.



Fig. 2 Residual concentrations and removal efficiencies of fluoride using different biomass dosing methods

Furthermore, removal efficiencies of fluoride that were lower under multi-contaminant condition could be attributed by the competition between fluoride and the kaolin particles (represented as turbidity) which are commonly negatively charged to adsorb on the available sites of the biomass. This finding indicates that fluoride removal was affected strongly by the presence of other contaminants in a system.

Under single-contaminant condition, the removal efficiencies of fluoride were higher because there was no interference with other elements in the system. Unfortunately, there is limited information to date regarding the removal of fluoride from multi-contaminant solutions to compare with.

3.3 Turbidity removal

It is obvious from Fig. 3 that MO alone could remove turbidity excellently with 96% (multi-contaminant solution) and 93% (single-contaminant solution) removals. Furthermore, MO proved its capability in improving the turbidity removal by BP from 65% (single-contaminant) and 43% (multi-contaminant) to the higher removals of 97% (single-contaminant) and 94% (multi-contaminant) when it was dosed sequentially after the treatment by BP.



Fig. 3 Residual concentrations and removal efficiencies of turbidity using different biomass dosing methods

Contrarily, the levels of turbidity were increased when BP was dosed sequentially after treatment by MO, indicating that BP was ineffective as turbidity removal agent. On the other hand, BP alone and combination of MO and BP (MO+BP) showed moderate removal of turbidity.

In terms of solution matrix, it is apparent that the removals of turbidity were higher in multi-contaminant solutions compared to single-contaminant solutions. The reason could be due to the interaction between electrostatic charges of heavy metals (positively charged) and the negatively charged of turbidity improved the removal efficiency.

4. Conclusions

Overall, this study demonstrates that it is necessary to investigate the removal behavior of target contaminants in different solution matrix. In this study, batch experiments were carried out under single- and multi-contaminant conditions. The results indicate that the removal behavior of target contaminants vary with different solution matrix.

There are several factors contributing to the variations in terms of removal behavior of one contaminant compared to another contaminant. For instance, the removals of lead, arsenic and turbidity were higher under multi-contaminant condition while the removal of fluoride was higher in single-contaminant condition. On the other hand, the removal behavior of nickel and cadmium were varied depending on the type of biomass dosing method.

From the results, it also can be suggested that the presence of more than one contaminant with different electrostatic charges under competitive condition (multi-contaminant) might suppress the retention of one contaminant on the biomass, thus reduced the removal efficiency. On the other hand, the removal of one contaminant does not have any interference with the other contaminants (i.e., fluoride), thus increased the removal efficiency under single-contaminant condition.

In terms of performance, combination of Moringa seeds and banana peels either dosed in mixing or sequential manner are seen can improve the treatment performance by single biomass for most of the contaminants. Therefore, this study is necessary to carefully investigate the variations in the removals of different target contaminants in order to ensure that the selected biomass were feasible to produce the anticipated removals in the actual environment. Further characterization studies on the biomass before and after the treatment process are recommended to be conducted in order to elucidate the mechanisms behind the removal of target contaminants.

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