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Potentiality for Removal of Heavy Metal from Groundwater Using Natural Adsorbents

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Abstract

Groundwater is one of the prime drinking water sources that are contaminated by human activities like the urbanization process, landfill leachate, using chemical fertilizer in agriculture, underground pipeline leakage, underground chemical, and petroleum products storage, etc. This study focuses on removing heavy metals from groundwater using natural adsorbents. Groundwater samples were collected from 16 different wards of Rajshahi City Corporation (RCC), where heavy metals, Manganese (Mn), and Iron (Fe), have been found extensively. As natural adsorbents, Azadirachta Indica (AI) leaf powder and Swietenia Mahagoni (SM) seed powder were used independently as well as in a combined ratio of 30:70, and 50:50 to treat groundwater. Different doses such as 150mg/L, 500mg/L, and 1000mg/L were selected with the contact times 5, 10, 15, 20, 30 minutes, 1, 2, 3, and 4 hours to determine the optimum doses and time. Study results showed that both the adsorbents removed 100% Fe from groundwater. The maximum removal of Mn was 60.47% in 30 minutes corresponding to 1000 mg/L of AI. Meanwhile, SM removed approximately 33.56% of Mn in 4 hrs when its dose was 150 mg/L. The combined effect of both AI and SM showed maximum removal of 28% corresponding to a mix ratio of 50:50. Overall, Azadirachta Indica seed powder performed better for removing heavy metals than Swietenia Mahagoni. The study provides information to the relevant authorities about the process parameters required for removing target percentages of pollutants from groundwater.

Keywords: Natural adsorbents; Groundwater treatment; Heavy metal; Pollutant removal percentage.

1 Introduction

Human beings are going towards a water crisis, because of its uneven distribution especially accelerated by climatic diversity, resulting in several wet and dry geographic zones, along with an acute increase in global freshwater requirement in current decades. In arid regions during draught season, surface water becomes unavailable and creates additional pressure on groundwater for irrigation, drinking, and other purposes. However, the limited groundwater resource is not always free from heavy metal pollution. Hazardous heavy metal pollution has been one of the most vital and sensitive environmental consequences, as it has become an influential threat to human health (Kim et al., 2020). Urbanization with a growing population and industrialization with growing technology along with human activities, generate a variety of heavy metal pollutants, especially in developing countries like Bangladesh.

Bangladesh extracts about 32 km³ of groundwater each year, 90% of which is used for agriculture and 10% for household and industrial uses together, accounting for 4% of worldwide groundwater extraction (Shamsudduha et al., 2020). In Rajshahi City and its suburbs, groundwater contamination has been identified as a serious issue. The key issue is the high iron concentration, which is in the range of 0.4-3.5 mg/L in the RCC region and 0.23-7.12 mg/L outside the RCC area, above the BD standard for drinking water requirements of 0.3-1.0 mg/L in both cases (Helal et al., 2011). Manganese concentrations were also elevated, varying from 0.1-1.52 mg/L in the RCC region to 0.23-2.40 mg/L outside the area, above the 0.1 mg/L drinking water standard limit mentioned by WHO (Helal et al., 2011). Total dissolved solids and arsenic levels across most locations were higher than the recommended level for potable water in Rajshahi City (Rasul & Jahan, 2010)

Several technologies are adopted for the treatment of water such as coagulation and flocculation, adsorption, chemical precipitation, ion exchange, membrane filtration, sand filtration, activated carbon adsorption, and up-flow anaerobic packed bed reactors (UAPB) (Shahi et al., 2020; Loloie et al., 2014). The performance of these methods varies with the types of coagulant used and the process parameters involved during water treatment. In addition, UAPB, ion exchange, activated carbon adsorption, membrane filtration, and chemical precipitation methods would be complex and expensive, particularly when treating a large amount of water/wastewater. These methods also produce toxic sludge that is harmful to humans and plants. These demands cost-effective, simple, and environment-friendly methods that can be used for the removal of heavy metals from groundwater.

Adsorption has been proven to be an effective method of drinking water treatment due to its flexibility in different aspects. Coagulants can be made from *Moringa Oleifera* (Chonde, 2017); Chitosan, Chitin, *Abelmoschus esculentus*, *Opuntia ficus-indica*, *Strychnospotatorum*, *Prosopis Laevigata* seed gum, *Hibiscus rosa-sinensis* (Awang & Aziz, 2012). AzaidSome other naturally occurring coagulants that are employed for wastewater treatment include *Sechium Edule* (Chayote), *Plantago Ovata*, *Zea mays* (maize), Cactus, *Tamarind Indica*, Bitter gourd seed, Cotton seed oil cake, Surjana seed powder, Maize seed powder, *Opuntia mucilage*, *Trigonella Foenum-graecus*, *Dolichos lablab*, Roselle seeds (Gautam & Saini, 2020). *Azadirachta Indica* can remove about 60% copper from wastewater in 60 minutes (Al Moharbi et al., 2020). *A. Indica* is used to remove heavy metals like lead (Pb^{2+}), chromium(Cr^{3+}), zinc (Zn^{2+}), cadmium (Cd^{2+}), nickel (Ni^{2+}), copper(Cu^{2+}), cobalt (Co^{2+}) and mercury (Hg^{2+}) from wastewater (Naseem et al., 2023). In another study, *Moringa Oleifera* removed 58% Cr, 70% Cd, and 65% Zn while *Tamarind Indica* removed 62% Cr, 73% Cd, and 70% Zn ((Prasad & Rao, n.d.). *Sweetenia Mahagoni* shell was found effective in removing chromium (IV). (Rangabhashiyam et al., 2016)

As natural adsorbents are used in water treatment and can exhibit high medicinal properties, this study uses these adsorbents to treat water. This study also finds the optimum doses and optimum time for each adsorbent to evaluate the maximum removal of heavy metals from groundwater.

2. Materials and Methods

2.1 Study area and sample collection

The study area is Sagorpara Bottola which is located on the border of wards 22 and 21 in Rajshahi City Corporation (RCC). The water sample was collected from a tubewell whose depth is approximately 100 ft. The tube well is being continuously used by the locals for drinking and other purposes since its formation. Water was drawn from the tube well for 3–4 minutes before taking the sample. The sample was then placed in previously cleaned and dried plastic bottles for storage. The water sample was then carefully transported to the lab immediately.



Figure 1. Sample Collection



Figure 2. AAS (Atomic Absorption Spectroscopy) machine

2.2 Selection of water quality parameters

The sample collected from Sagorpara was tested for the possibility of having heavy metals: Iron (Fe), Copper (Cu), Lead (Pb), Manganese (Mn), Arsenic (As), and Cadmium (Cd). The test was undertaken using AAS (Atomic Absorption Spectroscopy) machine in the Environmental Engineering Laboratory of Rajshahi University of Engineering and Technology (Figure 2).

2.3 Selection of natural coagulants

Based on the efficiency of pollutants removal from water or wastewater, two natural adsorbents have been selected. These are *Azadirachta Indica* (AI) leaf and *Swietenia Mahagoni* (SM) seed. Both of these plants are native to Bangladesh.

SM has many medicinal properties. Traditionally, it is used for malaria, hypertension, diabetes, and diarrhea, as an antipyretic, as a bitter tonic, and astringent (Bourdy et al., 2000). Pharmacological activities of SM are antimicrobial, anti-inflammatory, hepatoprotective, antidiarrheal, antiulcer, depressant, anticonvulsant and neuropharmacological, anti-diabetic, anti-HIV, immunomodulator, insect repellent and larvicidal, antifungal, antioxidant, analgesic, platelet aggregation inhibitors, antimutagenic and anticancer (Sukardiman & Ervina, 2020).

AI has been extensively used in Ayurveda, Unani, and Homoeopathic medicine and has become a cynosure of modern medicine. The pharmacological activities of the leaf of AI are Antifungal, Antibacterial, Antimalarial, Antifertility, Antipyretic, Anti-inflammatory, Analgesic, Antiulcerogenic, Antihypertensive, Antihyperglycaemic, Neuropharmacological, Antidermatophytic, Ororenal protection, Hepatoprotective, Immunostimulant, Antioxidant, Antigentotoxic, Anticancer (Subapriya & Nagini, 2005).

As AI and SM are both edible and have medicinal properties hence using them biologically to treat water is more beneficial compared to chemical treatment.

2.4 Preparation of natural coagulants

A good-quality AI leaf and SM seed were purchased from the Rajshahi City Corporation's closest local market. The shell that was covering the seed of SM was properly removed. After that, distilled water was used to clean the AI leaf and SM seed. Then these two adsorbents were adequately dried in the sun until the moisture is removed. A mortar and pestle were used to create a fine powder. The powders were sieved using a 600-micron stainless steel sieve before being employed as adsorbents (Figure 3).

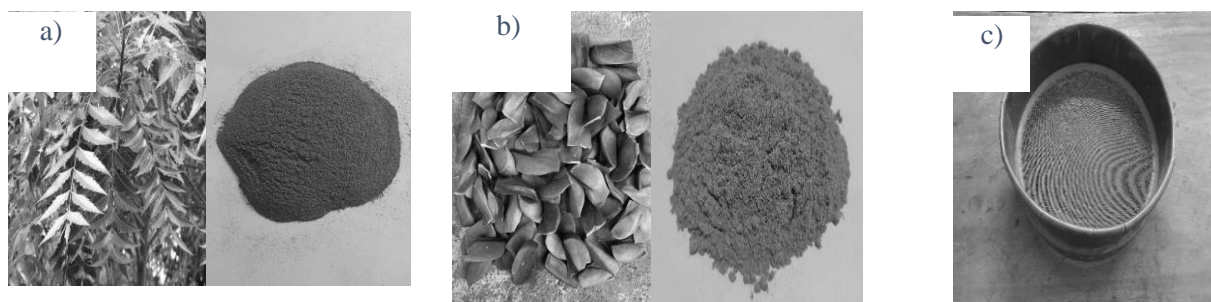


Figure 3. a) Azadirachta Indica Leaf and Powder, b) Swietenia Mahagoni Seed and Powder, c) 600 micrometer sieve

3. Results and Discussions

The groundwater from the tube well was found to be extremely contaminated by Manganese (Mn). The concentration of Mn found in the raw water is 2.53 mg/L, which is about 25.3 times more than the allowable limit of Mn in drinking water according to Bangladesh Standards. The concentration of Fe was found to be 1.85 mg/L which also exceeds the allowable limit of Fe in drinking water according to Bangladesh Standards. The other heavy metals in the raw sample were found to be zero or way below their allowable limit in drinking water.

Both the adsorbents alone and their combined doses were found very effective for removing 100% of Fe from the groundwater in 5 minutes. The removal of Mn varied for different adsorbents and their combined effect. So, the variation in the removal of Mn due to the effect of natural adsorbents is mainly discussed here.

3.1 Effect of AI

Figure 4 shows the removal percentage of Mn due to the effect of AI for up to 4 hours in an intermittent manner. The adsorption process was conducted for 3 doses of AI: 150mg/L, 500mg/L, and 1000 mg/L. The adsorption of Mn by AI is seen irregularly with time though each dose causes a significant reduction in the concentration of Mn in raw water.

For a dose of 150 mg/L, after 5 min the removal percentage due to the effect of AI was about 14.5. The adsorption pattern shows irregular drops and a rise in Mn concentration afterward. After 10 min of adsorption, the concentration of Mn drops until rising again after 15 minutes. After 4 hours, AI was able to remove 31.225% Mn from the raw water (Figure 4).

When the dose of AI is increased to 500mg/L, the removal of Mn also increased. After 5 minutes, the Ai was able to remove about 28% Mn from groundwater. The adsorption pattern is also irregular here. The removal percentage increases up to 30 min and then drops at 1 hour and 2 hours. After that, the removal percentage again rises at 3 hours and then again drops at 4 hours. After 4 hours the removal percentage of Mn became 46.957. (Figure 4)

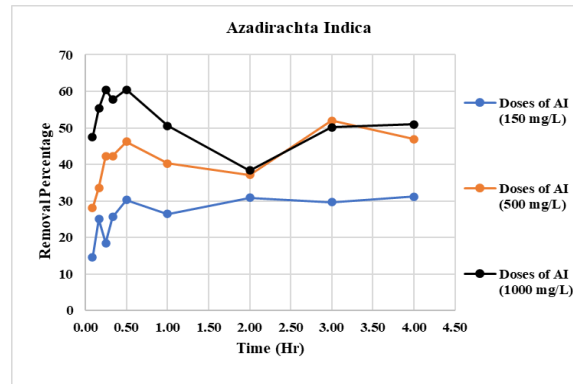


Figure 4. Removal percentage of Mn with time due to AI

The dose of AI was further increased to 1000mg/L which gives more removal of Mn than other doses of AI. After 5 minutes, AI removed about 47% of Mn from the raw water. After 4 hours the removal percentage of Mn due to the effect of AI became 51 (Figure 4).

AI exhibits carboxylic and hydroxyl groups which are involved in different hydrostatic reactions to remove heavy metals from water (Naseem et al., 2022). The adsorption capacity of AI may depend on pH, temperature, particle size of adsorbents, adsorbent concentration, doses, etc. While most of these parameters are controlled, due to the temperature change in the atmosphere as well as the media, the irregular adsorption pattern by AI was likely to be seen.

The maximum removal percentage of Mn was 60.47 in 30 minutes when the doses of AI were 1000mg/L. So, this dose and time can be considered the optimum dose and time for AI in the removal of Mn from groundwater.

3.2 Effect of SM

Figure 5 depicts the intermittent removal percentage of Mn caused by the impact of SM for up to 4 hours. Three dosages of SM were used in the adsorption process: 150 mg/L, 500 mg/L, and 1000 mg/L. Even though each treatment significantly lowers the amount of Mn in water from what was initially in raw water, the adsorption of Mn by SM is seen to be uneven over time.

For dose 150mg/L, after 5 min the removal percentage of Mn due to the effect of SM became 23.24. The adsorption pattern was irregular due to the temperature changes. The initial removal percentage of Mn decreased up to 20 minutes. After that, the removal percentage showed an increasing trend up to 4 hours. After 4 hours, the removal percentage of Mn became 33.56 (Figure 5).

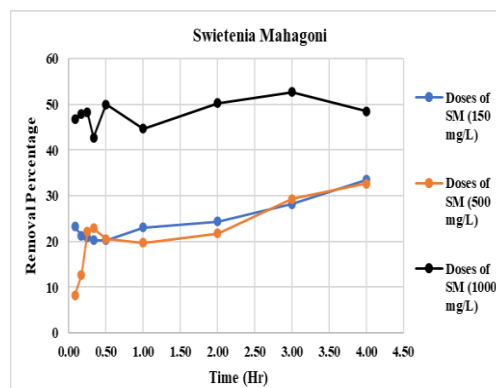


Figure 5. Removal percentage of Mn with time due to SM

When the dose of SM is increased to 500mg/L, the removal of Mn is found minimum after 5 minutes with the removal percentage being only 8.3%. As time progressed, the adsorption pattern shows irregular drops and a rise in Mn concentration. After 4 hours the removal percentage of Mn became 32.65.

The dose of SM is further increased to 1000mg/L which gives more removal of Mn than the other 2 previously selected doses. After 5 minutes, the SM removed about 46.88% Mn from raw water. Initially, the removal pattern showed some drops and rises. After 3 hours, the SM showed maximum removal capacity which is about 52.69%. After 4 hours, the removal percentage drops down to 48%. (Figure 5)

The maximum removal percentage of Mn was 52.69 in 3 hours when the dose of SM was 1000mg/L. So, this dose and time can be considered the optimum dose and time for SM in the removal of Mn from groundwater.

3.3 Combined effect of AI and SM

In this phase, the combined effect of natural adsorbents is evaluated. The dose is kept constant which is 150mg/L and, SM and AI are added in a 70:30 and 50:50 ratio. The intermittent removal percentage of Mn caused by the combined impact of SM and AI are determined for up to 4 hours (Figure 6). Even though each treatment significantly lowers the amount of Mn from raw water, the adsorption of Mn by SM and AI is seen to be uneven over time.

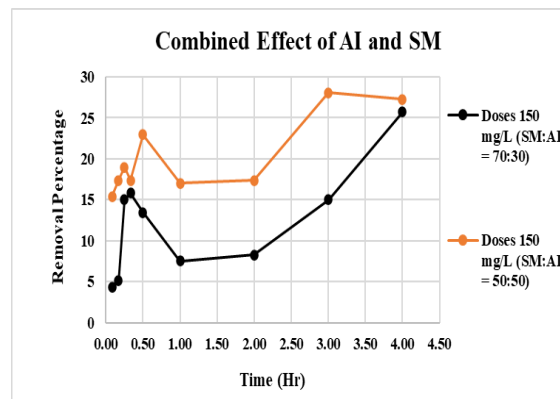


Figure 6. Removal percentage of Mn with time due to the combined effect of SM and AI

When the ratio of SM and AI is 70 to 30, the removal of Mn from raw water is 4.35%. As time progressed, the adsorption pattern shows irregular drops and a rise in Mn concentration. The adsorption pattern rises for up to 20 minutes and falls after that for up to 1 hour. After 1 hour, the pattern of removal of Mn shows an increasing trend. At 3 hours, the removal rate of Mn becomes a maximum of 28%. After 4 hours the removal percentage of Mn became 25.69 (Figure 6).

When the ratio of SM and AI is 50 to 50, the removal percentage of Mn from raw water was 15.42. As time progressed, the adsorption pattern shows irregular drops and a rise in Mn concentration. Mn concentration drops after 4 hours, resulting in a removal percentage of Mn being 27.27.

Between these two ratios, the overall removal percentage of Mn is comparatively better when the ratio of SM and AI is 50:50. The maximum removal rate is 28% after 3 hours when the ratio of AI and SM was 50:50.

Conclusion

The study will assist to determine how easily and affordably natural adsorbents can be utilized to treat groundwater. Both the adsorbents and their combination have removed 100% of Fe from the groundwater in 5 minutes. The removal percentage increases with increasing doses of AI. The maximum removal percentage of Mn was 60.47 in 30 minutes when the doses of AI were 1000mg/L. The maximum removal percentage of Mn was 52.69 in 3 hours when the dose of SM was 1000mg/L. These times and doses can be considered optimum times and doses for respective coagulants. AI is more efficient for the treatment of groundwater. For the case of the combined effect of SM and AI, the overall removal percentage of Mn is comparatively better when the ratio of SM and AI is 50:50.

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