Assessing plant-based coagulant performance to improve the safety and quality of water
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ABSTRACT

The proposed work was carried out to replace the conventional water purification methods by natural coagulants. Samples of groundwater were obtained and analyzed for quality parameters. The coagulants of Moringa oleifera, orange peels, and date pits were prepared, and different combinations of these coagulants were used to find out the most effective one. Results regarding various physicochemical tests including pH, electrical conductivity, turbidity, total dissolved solids (TDS), calcium, magnesium, total hardness, bicarbonates, chlorides and heavy metals revealed that pH was not affected by treatments, values for EC were dropped down slightly but not reduced to an acceptable limit of World Health Organization (WHO). TDS, total hardness and chlorides reduced to the acceptable limit of WHO. Additionally, a significant reduction was recorded in lead and arsenic contents of treated water samples. Moreover, the results exhibited that the values for TPC and Total Coliform were dropped down slightly but not reduced to a satisfactory limit of WHO, while fecal Coliform reduced to the acceptable limit of WHO and E.coli was found to be zero in all samples. Conclusively, natural coagulants contain good adsorbent properties, which can confer the purification of water in a cost-effective way.


1. Introduction

Water is universal solvent as it dissolves more substances than any other liquid, because of its hydrogen bonding. Water has several important functions in climate system, progressions of life, daily hygiene, sanitation, food processing and several other uses that without it, life would be impossible. Almost 80% of world’s population is suffering from shortage of safe drinking water (1). Approximately 17% of the world's population uses water from the contaminated sources, 32% population is consuming water from secured sources and 51% of people are obtaining water from wells or ground water (2).

About 20% of Pakistan’s population has access to safe drinking water, fit for human consumption (3). The several factors which contribute to water contamination may include industrial wastes, agricultural wastes, deforestation, pollution and over population. Due to the effect of anthropogenic contamination of ground water, a rise of 200 percent is observed in patients (4). Contaminated water can cause a lot of health problems in human beings by increasing the rate of disease risk and mortality which is high in children under five years, pregnant women and older people. In Pakistan, mostly people rely on underground water or municipal water supply for drinking purpose (4). Different types of constituents which include physical, chemical and microbial contamination may contribute...
to water pollution in different ways. Pathogens are disease-causing organisms that can contribute to microbial contamination and may come through fecal contamination which can cause many diseases like diarrhea, cholera, typhoid and dysentery (5). Chemical contamination may include minerals that are non-harmful contaminants, but they can influence the sensory characteristics of water like taste, smell, color or temperature of water, and make it unacceptable to the public. Heavy metal contamination can cause severe problem in human beings. Among them arsenic is the most chronic and causing worldwide health issue like skin lesions, hypertension, neurological effects, vascular disease, respiratory disease, diabetes mellitus, and skin cancer. Skin is very sensitive to arsenic and its early symptoms include skin lesions even in the range of 0.005-0.01 mg/l (6). Chemicals may also include agro chemical wastes like fertilizers, weeds and burnt crops that are the major reasons of water pollution (7). Contaminated water also contains pathogens which are disease-causing organisms and comprise of bacteria, amoebas and viruses etc. Chemical and microbial water contamination is occurred due to improper management of industrial and human wastes and the consequences lead to different health problems after consumption (8). Many conventional methods are being used to treat drinking water like boiling, chemical treatments, ionization, ozonation, home filtration. According to WHO recommendations, boiling could be a good method to kill or inactivate the microbes in water which can cause diarrhea. In a situation of emergency, boiling was the most ideal approach to purify water that is also risky as protozoan parasites, microorganisms or infections could not be removed completely (9). Conventionally, chemical treatment was mostly done through chlorination and coagulation- flocculation which can also cause toxicity in case of toxicity from metal ions (45). Chlorine used for disinfection made the pathogens inactive and provide a barrier against recontamination (10). Additionally, this method could be used at household level (11). On the other side, it is not good in decreasing physical and chemical contamination because water disinfected with chlorine may have residual chlorine which is carcinogenic and could cause heart attack as well (12). In ionization UV rays and ionizing rays (x-rays, gamma rays and electrons beam) are used to purify water commercially, but the black side is that some organisms are resistant to UV light which includes cryptosporidia (13). Ozonation is also being extensively used for water decontamination as ozone can kill microorganisms and treat organic matter efficiently (14). Water treatment through reverse osmosis is another useful technique which has been effectively used for the last 40 years and proved to be effective for removal of contaminants and microbes (15). Pesticides are hazardous to human health causing many diseases which can also be removed through reverse osmosis and nano filtration (16). Home filtration is currently being used at household level as it does not need any further disinfection to purify water. Major drawback of this approach is its cost, because if filter bed is not replaced intermittently, microbes may proliferate and can cause contamination.

All over the world, research has been focused in utilizing the herbal waste materials and to rationalize waste disposal techniques by making it more economical. They are also characterized by environment friendly behavior and claimed to be secure for human health. Organic coagulants are natural polymers obtained from renewable resources which are environmentally friendly and safe to human health (17). Recent studies show that Moringa oleifera, maize mesquite, cactus latifaria, melon seed husk, neem leaves, luffa cylindrical extract, orange peel and date pits can be utilized for such purposes (18). Moringa seed powder has antimicrobial activities which helps in lowering the bacterial count (19). Moringa seed powder is reported to reduce the hardness and turbidity of water up to 60% and 99% respectively and is being widely used for water treatment in many developing countries. It can remove hardness up to 60% as well as 99% turbidity and it is successfully being used in many developing countries as a mean of water purification. The protein content of Moringa Oleifera used in different concentrations can lower down the pathogenic level of water. Protein fractions include globulin and albumin, globulin imparts bactericidal effect to kill E. coli while albumin shows bacteriostatic effect against E. coli (44). Fruit peels are highly effective in the removal of chemical contaminants and heavy metals. Several fruit peels are easily available at local markets at low cost and can be used as alternative adsorbent to treat water (20). The main components of orange peel are cellulose, pectin, hemicelluloses, lignin, and other low molecular weight compounds. Orange peel is considered as good absorbent as it is abundantly present in nature with no toxic effects and having easy access to its biodegradability. Another agricultural based waste material includes date pits and its
disposal is a major problem for date industry. Date pit obtained from date palm is the good source of activated carbon which is being used as the source for water purification during the last few decades (21). Accordingly, the current project is designed with the aim to find out a new, easy, low cost and effective method of water purification from indigenous resources and to identify the best use of plant-based extracts having high coagulating properties for reducing the microbial activities in water and making it drinkable without any health hazards.

2. Materials and methods

2.1. Collection and preparation of natural coagulants

Samples were collected in sterile bottles after sterilizing the tap with spirit lamp to avoid contamination. The bottles were labeled with tags containing time, date, area, sample code and transferred to the laboratory immediately for analysis. Moringa oleifera seeds (Suhanjna), Phoenix dactylifera (Date pit) and Citrus sinensis (Orange peels) were manually cleaned to remove damaged seeds, dust particles, seeds of other crops and then washed. After that oven drying was done at 50-60°C for 24 h and then material was ground into fine powder using food grinder. Seeds were sieved using a strainer to obtain a fine powder. The end product obtained after sieving was referred to as natural coagulants (21).

2.2. Sample preparation by Jar test method

The jar test method was followed in this study. Raw sample was taken in a beaker of one-liter volume. Two grams of coagulant Moringa oleifera was added to a beaker and mixed vigorously for 2 min followed by 15 min of gentle mixing. The suspension was allowed to stand still for a period of 1.5 h. Finally, the supernatant was centrifuged and then filtered through Whatman filter paper No.4 for further analysis. Similar procedure was carried out for other coagulants like, Orange peel, date pit and their combinations (18).

2.3. Experimental set-up

Water samples were checked for contamination, different trials were made to check the best dose and significant results were found at 2 g of following plant materials. Seven treatments which were used in this study are T° as raw water (control), T1 as Moringa oleifera powder (MO-100%), T2 as Date pit powder (DP-100%), T3 as Orange peels powder (OP-100%), T4 as Moringa oleifera powder + date pit powder (MO + DP-50%+50%), T5 as Moringa oleifera powder + Orange peel powder (MO + OP-50%+50%), T6 as Date pit powder + Orange peel powder (DP+OP-50%+50%), T7 as Moringa oleifera + Date pit powder + Orange peel powder (MO + DP + OP- 33.33%+33.33% +33.33%).

2.4. Chemical analysis

All the water samples were checked for pH, EC, turbidity, TDS, minerals (calcium, magnesium, chlorides), total hardness, bicarbonates and heavy metals (lead, arsenic). All analysis was carried out according to the methods prescribed in APHA. (22)

2.5. Microbial Analysis

2.5.1. TPC (Total plate count)

Total plate count in the water sample was checked by (23) by using media named as plate count agar.

2.5.2. Coliform count and E.coli

Membrane filtration method was used to detect coliform and E.coli by the method of ISO 9308-1:2000. Oxidase test was used to confirm the colonies of coliform and Kovac’s reagent was used for the confirmation of E.coli.

2.5.3. Fecal coliform

The same procedure was followed for fecal coliform determination as used for total coliform determination. Colonies having blue colour were confirmed as fecal coliforms while colonies having grey or creamy colour were observed as non fecal coliforms.

2.6. Sensory evaluation

Sensory parameters include color, odor and taste, raw samples were checked by using 9 point hedonic scale which was assessed by different people (24). The acceptance level of consumers could also be accessed through this process (25).

2.7. Statistical analysis

The data obtained from 7 treatments were analyzed to determine the level of significance and ANOVA will be used to obtain the difference in the means of values under CRD (Completely Randomized Design) to check out the effect of various treatments on quality and safety parameters of water and to select the most appropriate treatment. Microsoft Excel sheet was used to compile the data in graphical form and to develop association between the results.
3. Results

Three organic matters were used to make coagulants by jar test procedure, which were used to treat contaminated water. Moreover, seven treatments were made and then applied to the sample and their results were compared with WHO limits.

3.1. Physicochemical analysis

3.1.1. Effectiveness of treatments on pH of water

The pH of ground water is affected by soil composition, organic matters in water and by the addition of many chemicals in water bodies. According to WHO the acceptable limit of pH values for drinking water is 6.5-8.5. From the results shown in figure 1, it is concluded that pH of all the treatments were within the acceptable limit of WHO, while there is a rapid increase in pH value of T6 (Date pit powder + Orange peel powder) and T7 (Moringa oleifera + date pit powder + Orange peel powder).

3.1.2. Effectiveness of treatments on turbidity

Turbidity is defined as the intensity of light to be scattered. If the intensity of scattered light is higher it means water has higher turbidity values and vice versa. The permissible limit of turbidity for drinking water set by WHO is 5 NTU. It is deduced that the turbidity value for all the water treated or untreated was zero, it means that no turbidity exists in the untreated and treated water samples. All the treated water samples are within the limit of WHO which is 5 NTU.

3.1.3. Effect of coagulants on Electrical Conductivity of water

Figure 1 shows that the higher mean value of EC was observed for T0 (raw sample), while the lowest mean value was observed for T4 (MO+DP). From the results of all treated water samples it is deduced that EC for treated water samples was decreased this is because of the presence of lower molecular proteins present in them.

3.1.4. Effectiveness of herbs on Total Dissolved Solids

The highest mean value was recorded for control (raw sample) while the lowest mean value was observed for T7 (Moringa oleifera + date pit powder + Orange peel powder). TDS of water samples decreases after all treatments moreover; all water samples are within the limits of WHO. From figure 1 it is concluded that the highest mean value was recorded for control (raw sample) while the lowest mean value was observed for T7 (Moringa oleifera + date pit powder + Orange peel powder). TDS of water samples decreases after all treatments because of the presence of lower molecular weight soluble proteins which carry positive charge.

3.1.5. Effectiveness of herbal coagulant on hardness of water

About 10-500 mg/l total hardness of water is allowed by WHO. Water is classified into different categories according to the concentration of calcium carbonate. Water will fall in the category of soft water, if water has concentration of 0-60 mg/l of CaCO₃ as hardness of water. 61-120 mg/l of CaCO₃ will come under the category of moderately hard, 121-180 mg/l of CaCO₃ will be considered as hard water and water having 181 mg/l of CaCO₃ is considered as very hard water.

According to figure 1, all the water samples were within the limits of WHO. The highest mean value was recorded for control (raw) sample and the lowest mean value was observed for T1 (Moringa oleifera powder). The main reason for lowering of hardness of water is the coagulating effect of active elements present in natural materials having cationic proteins with lower molecular weights.

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3.2. Mineral analysis

3.2.1. Calcium
From the figure 2, it is deduced that the highest mean value was recorded for T3 (Orange peels powder) because orange peels are the rich source of calcium and the lowest mean value was observed for control sample. All the water samples were below the acceptable limits except for T2 (Date pit powder), T3 (Orange peels powder) and T5 (Moringa oleifera powder + Orange peel powder); this is because orange peels and date pits are a rich source of calcium. The WHO recommended value for calcium is 70-200 mg/l.

3.2.2. Magnesium
Figure 2 represents the graphical representation of all treatments of water samples. The highest mean values were recorded for control sample while the lowest mean value Mg was recorded as T3 (Orange peels powder). All the recorded value was according to the permissible limits of WHO. The permissible limit of magnesium according to WHO is 50-150 mg/l.

3.2.3. Chlorides
From the results shown in figure 2, the highest mean value was observed for raw water sample which was exceeding from the limit set by WHO and the lowest mean value was observed for T7 (Moringa oleifera + date pit powder + Orange peel powder). All the treatments were effective for lowering of chlorides in water samples. The recommended value of chlorides in water is about 250 mg/l.

3.3. Toxic metal analysis

3.3.1. Arsenic
The permissible limit of arsenic is 10 µg/l as declared by WHO. The figure 3 shows the results of arsenic in the water samples, the highest mean value was observed for T0 (Raw sample) and lowest value of arsenic was observed for T4 (Moringa oleifera powder + date pit powder) sample. All the water samples showed higher arsenic values than the acceptable value set by WHO while all treatments were proved effective in lowering the arsenic contents in water sample. The reason behind this heterogeneous property is that the organic matters contains low molecular weight amino acids which contains active group of binding agents which are physiologically active in nature and could bind the metal ions.

3.3.2. Lead
Figure 3 shows the highest mean value was observed for T7 (Moringa oleifera + date pit powder + Orange peel powder) sample which was exceeding from the limit set by WHO and lead was not detected in T1 (Moringa oleifera powder), T2 (Date pit powder), T3 (Orange peels powder) and T4 (Moringa oleifera powder + date pit powder) samples, which means these treatments were effective for lowering of lead in water samples. According to WHO the acceptable limit of lead is about 0.01 mg/l.
3.4. Microbial analysis

3.4.1. Efficiency of herbs in lowering Total Plate Count (TPC)

Figure 4 shows the mean values for TPC of samples which were treated with Moringa oleifera, date pits, orange peels and their combinations. From the results it is inferred that the highest mean value was observed for untreated samples while the lowest mean value was observed for T4 (Moringa oleifera powder + date pit powder) for which the value was recorded as 2.1 x 10^2 ± 3.05. All the samples were crossing the acceptable limits but after treatment microbial load lowers down.

3.4.2. Efficiency of herbs in lowering Total Coliform Count

It can be noticed in figure 4, that higher mean value was recorded for raw sample and the smallest mean value was recorded for T4 (Moringa oleifera powder + date pit powder). The value for T4 was recoded to be 3.4 x 10^2 ± 6.4. All the values were exceeding the WHO limits but the treatments lowered down the count of Total coliform.

3.4.3. Efficiency of herbs in lowering Fecal Coliform

The fecal coliform should not be more than 1 cfu/250 ml. The fecal coliform count was observed positive in raw water samples which was 2.43 x 10^2 ± 3, while in treatments T1, T2, T3, T4, T5, T6 and T7, fecal coliform count was found as negative. The results show that all treatments were effective for the removal of fecal coliform count and all the results are within the limits of WHO.

3.4.4. Efficiency of herbs in lowering E.coli numbers

The count for E.coli was observed to be negative in raw water samples as well as treated water samples. The results are found to be with in the acceptable limits of WHO.

3.5. Sensory analysis

Figure 5 specifies the results obtained from 9 point hedonic scale for all sensory parameters which includes taste, odor, color and overall acceptability. All the scores were less than or equal to 5 and according to 9 point hedonic scale, score 5 indicates that sample is neither liked nor disliked by consumers.

4. Discussion

Human health is not directly affected by pH while some indirect association has been observed in the past studies like change in pH allows the microbial survival as well as increases the solubility of metals which results in gastrointestinal problems in human beings (29). Current results for pH agreed with the
previous research in which effect of Moringa oleifera was examined on quality of water samples obtained from different areas of Faisalabad and found no significant effect on pH of water (30). Turbidity means the presence of haziness or any cloudiness in water, which is described as the optical character of water and measured by Nephelometric Turbidity Unit (NTU). It affects the color as well as quality of water by contaminating the water with suspended particles like soil particles or microbes. From the previous study it is revealed that date pits could remove turbidity up to 95% (31). In another study it is revealed that orange peels act as a natural coagulant (32). In a study conducted about Moringa oleifera water clarification it was found that moringa was effective in the removal of turbidity of treated water(33).

The acceptable limit of TDS is 1000 mg/l and water having more than 1000 mg/l TDS will impart a taste to drinking water while water having TDS less than this range is considered as fresh water. Current investigation is in agreement to the previous study in which orange peels and Moringa oleifera was effective to decrease total dissolved solids in water (18). In another study moringa olifornia was used for the water treatment and found to be effective (35). Electrical Conductivity is the ability to measure dissolved ionic content in water. EC has a direct relationship with ionic contents of water, the increase in EC means increase in ionic contents of water and vice versa. It is measured in Siemen (µS/cm). High values of EC could cause increase in blood pressure which can affect kidneys also. Current investigation was in agreement of the research conducted to reduce pH, EC and heavy metals in waste water with the help of Moringa oleifera treatment (34). Hardness is due to the mineral contents of water specially calcium and magnesium and their sum are considered as hardness of water. The natural source of hardness is CaCO₃ or MgCO₃ while water having CaCO₃ hardness in the range of 15 to 375 mg/l is considered as fresh water. The study agreed with the previous results and her research work by Muyibi and Evison in 1995 in which Moringa oleifera was effective for the softening of water. In another study it was deduced that moringa was effective for the removal of hardness upto 70% while orange peels could remove hardness upto 65% (18). Ca is an important mineral for human health, without it many health problems could occur which includes osteoporosis, diabetes and cardiovascular health problems. In a study was found that orange peels was a rich source of calcium that is the main reason of increasing calcium levels in water (37). According to findings of WHO a meeting was conducted in Italy, Rome in 2003 in which proposal was given that magnesium was also important in drinking water and should be a part of drinking water because it also have some health effects (38). The current study was in agreement of the previous study in which it was observed that orange peels was 80% effective for lowering of Mg (18). Chlorides give salty taste to water and when water is passed through pipes, chlorides may react with pipe by increasing the level of metal. Current study agreed with the previous findings that with Moringa oleifera and orange peels were effective for lowering the level of chlorides (18).

Arsenic is injurious to human health because of its carcinogenic effects on both animals and human beings leading to cancer, nervous problems, cardiovascular diseases, diabetes and hypertension. A huge population is at the risk of arsenic contamination that every 10th people in millions are at risk (39). The current study was in agreement of the previous study in which water was treated with Moringa oleifera and found that moringa proved to be beneficial in lowering the level of arsenic in water (40).

Lead is symbolized as Pb and its atomic number is 82. Corrosive pipes could be the cause of lead contamination in water. Lead may adversely affect the digestive system, nervous system and reproductive system of human beings. Moreover, it can cause learning and memory issues in people. The current study was in agreement with the previous study in which moringa seeds and date pits proved to lower the lead contents in water samples (41). The current results also matched with the previous study in which orange peels played a vital role in lowering the lead contents of water samples (42). Findings of the current study are in agreement of the previous study in which water was treated with Moringa oleifera and found that total bacterial count lowered down efficiently (26). The current investigation follows the pervious one in which total coliform counts were lowered down by treatment of water with Moringa oleifera (26). The current results showed harmony with the previous findings in which fecal coliform count was lowered down by treatment of water with Moringa oleifera (26). The outcomes of the current research are in line with the earlier research in which E.coli was lowered down by treatment of water with Moringa oleifera (27).
Another study was performed to check out the effect of orange peels to lower down the microbial activity of *E.coli* in water and found that orange peels could lower down *E.coli* from $10^3$ to $10^2$ cfu/g (28).

5. Conclusion
In the current study, the herbs (Moringa oleifera, date pits and orange peels) were evaluated as a natural coagulant to remove contaminants from water. The study concludes that chemical parameters of water improved to an extent after all treatments, especially for T4 (Moringa oleifera and date pits) which included the combination of Moringa oleifera and Date pits. It also reveals beneficial in lowering the count of heavy metals which are toxic to human health. Moreover, all the treatments indicated to be effective in lowering the bacterial count. These herbs can act as disinfectant the water to reduce the contamination and make it drinkable at local level by removing the suspended and dissolved particles. However, they did not guarantee the removal of pathogens completely. Highly significant changes were observed in color, odor and taste of treated water samples which were overall not accepted by the consumer. Further studies in this regard are needed to improve the overall sensory characteristics of water and to ensure the removal of pathogens and make it completely drinkable.

Conflict of interest
The authors declare no conflict of interest.

Acknowledgement
The authors are grateful to National Institute of Food Science and Technology, Faculty of Food Nutrition and Home Sciences, University of Agriculture and Science and Technology, Faculty of Food Nutrition and Home Sciences, University of Agriculture and Science and Technology, Faisalabad for performing the experiments and preparation of the manuscript.

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