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Treatment of wastewater using modified Bentonite and its impact on soil and properties

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Abstract: Nowadays, it's normal practice to use treated wastewater as a substitute water supply for irrigation. One of the most efficient and cost-effective techniques with the greatest potential for removing and recovering pollutants from wastewater is the adsorption process. In this study the impact of Bentonite (Ben) and Bentonite-HA complex on wastewater treatment were evaluated. The treatment of wastewater was carried out through the adsorption onto Bentonite and Bentonite-HA complex using fixed bed column technique. The treatment carried out with flow rate 200 and 250 ml /H for each 10 gm Bentonite and Bentonite-HA complex, respectively. The results clarified that TOC was completely removed from WW and removed by a 97.7% ratio by Bentonite-HA complex and Bentonite, respectively. In addition, Iron, zinc and lead were completely removed from the WW by Bentonite -HA complex. Moreover, other heavy metals (cobalt, manganese, cadmium, chromium, copper and Boron), total dissolved solids (TDS), EC, and pH of the treated wastewater (TWW) by then Bentonite and Bentonite-HA complex were within the acceptable range of irrigation. Hence this study indicated the treated wastewater by Ben_HA complex much beter than Bentonite in wastewater treatment. The impact of irrigation with wastewater and treated wastewater on the chemical characteristics of the soil and faba beans was investigated in a pot experiment. Analysis of plants and soil revealed that irrigation with treated wastewater significantly reduced heavy metals, total organic carbon, EC, total dissolved solids, and pH, resulting in substantial changes in plant and soil attributes compared to untreated irrigation. This study's intriguing conclusion is that treated wastewater can be regarded as a useful irrigation supply. Reusing treated wastewater in agricultural systems once it has undergone complete treatment is encouraged.

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Key words: Bentonite, COD, HA, wastewater, Agriculture, Faba Bean, Soil, Adsorption.

1. Introduction

One of the biggest environmental issues facing the globe today is the contamination of water supplies by harmful metal species. Because of its harmful effects on humans and plants, heavy metals as Cd^{2+}, Fe^{2+}, Zn^{2+}, and pb^{2+} have recently raised concerns about water contamination [(1) (2)]. Because these metal ions build up in food chains, their presence in water puts aquatic life and humans at risk.

Nowadays, several processes have been used to remove inorganic and organic pollutants of different sources from water, including membrane filtration, adsorption, coagulation and flocculation [(3)]. Using adsorbents materials such as biochar and clay minerals are those recently in the purification of wastewater by removing heavy metals, organic and inorganic contaminants. [(4)]. As well as, due to its exceptional adsorption efficiency, commercially available activated carbon is the most popular and efficient adsorbent [(5)]. Furthermore, due to their high specific surface area, chemical and mechanical stability, a range of surface and structural characteristics, and affordability, clay minerals such as montmorillonite, kaolinite, illite, and bentonite are frequently utilized for wastewater treatment [(6)].

For Cd, Cr, Co, Cu, Fe, Pb, Mn, Ni, B, As, and zinc in their ionic forms from aqueous media, clays have drawn notice as superior adsorbents. Additionally, the type of clay utilized affects the adsorption capabilities, which vary from metal to metal [(7) (8)].

A net negative surface charge is produced on the clay by the isomorphous substitution of Al^{3+} for Si^{4+} in the tetrahedral layer and Mg^{2+} or Zn^{2+} for Al^{3+} in the octahedral layer. Bentonite is a 2:1 type of clay, meaning that the mineral is made up of units of two silica tetrahedral sheets with a central alumina octahedral sheet [(9)(10)]. Bentonite has superior sorption qualities when compared to other forms of clay. It has sorption sites on its outer surface, edges, and interlayer space [(11)]. Because of its physical and chemical characteristics, bentonite is regarded as one of the most promising options for the decontamination of high-level heavy metals. [(12)]

One of the main ingredients of humic substances, which are produced when microorganisms break down biomolecules, is humic acid (HA). HA may bind a variety of contaminants, such as hazardous heavy metals and synthetic organic compounds, and transport them through water treatment plants and distribution networks [13]. HA's structure causes it to function as a polyelectrolyte in solution. Adsorption of HA molecules on the bentonite-HA complex surface hence increases the metal cations' adsorption capacity due to HA's potent complexing sites [14] [10].

Thus, the present study demonstrates the removal of heavy metals and organic contaminants by using Bentonite and Bentonite-HA complex from wastewater was utilized as a low-cost adsorbent and economic method for water treatment. As well as, evaluate the using of treated wastewater in irrigation and its impact of soil and plant properties.

2. Materials and Methods

2.1. Wastewater sampling

Sewage effluent was gathered from the Eldare region of Egypt's Sohag province. Wastewater was thoroughly chemically analyzed to identify its EC, pH, total organic carbon (TOC), heavy metals, pathogens, and major cations and anions both before and after the treatment procedure.

2.2. Bentonite

With a high specific surface area, cation exchange capacity, and adsorptive attraction for both organic and inorganic ions, bentonite is a mineral that is frequently found in large quantities in soils. For this work, bentonite was acquired from the Egyptian Petroleum Research Center's source clay minerals repository. The bentonite fraction was sieved into particles with a diameter of 80 nm.

2.3. Preparation of Bentonite-HA stock

Getting Bentonite and HA Prepared For the adsorption experiment, stock materials, bentonite suspensions (25 gram) stock solutions, were set at pH 6 and 0.001M KCl. The source of HA in its potassium salt was Sigma-Aldrich Co. in Germany. In a 1000 mL beaker, distilled water and HA powder were combined. To improve the solid-state HA's dissolution; a few drops of KOH (0.1 M) were added while stirring. The pH was then adjusted to 6 by adding sodium hydroxide or hydrochloric acid, and the ionic strength of HA solutions was adjusted to 0.001 M by adding KCl salt. The concentration of HA in the stock solution was 0.5 g L^{-1}. At 4 °C, the prepared solution was kept. [15] [16]

2.4. Bentonite and Bentonite -HA complex Characterization

(1) SEM analysis: The scanning electron microscopy (SEM) was used to explore the Bentonite morphology.

(2) BET Surface area: The method of Brunauer, Emmett and Teller (BET) are regularly applied to estimate the materials surface area.

(3) FT-IR spectroscopy, FT-IR spectra of Bentonite and Bentonite–HA complex have been studied. Significant understanding of the reactivity, structural organization of the functional groups, and aromatic/aliphatic domains in materials has been determined by FT-IR spectroscopy.

(4) Particle size distribution: Particle size distribution was used to investigate the particle size of a Bentonite and Bentonite –HA complex

(5) Zeta potential: is an indicator to the stability of colloidal dispersion**.**

2.5. Treatment of wastewater by adsorption on Bentonite and Bentonite -HA complex

Treatment procedure was done with Bentonite and Bentonite -HA complex by using fixed bed column model. The Column has a diameter of 3cm and 30 cm length. The rate of adsorption process was detected every day.

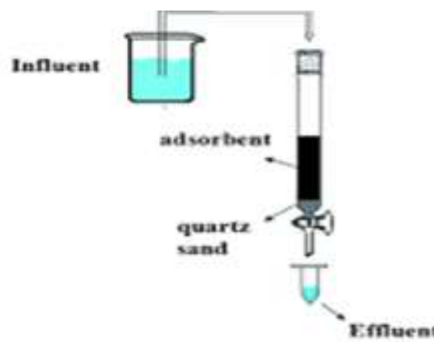

Fig. (1) Fixed bed column model

2.6. Physical and chemical analysis of soil

The mechanical analysis was used to determine the soil texture. The mechanical analysis of soil sample was carried out using pipette method as described in reference by (**Morteza Mohsenipour et al., 2012)** [17] [18].

2.6.1 pH value

The soil pH has great importance regarding to the soil properties. The acidity (hydrogen ion concentration) was determined by means of digital pH meter (cole parmer) in suspension (1 soil: 2.5 water) after 15 minutes of orbital shacking [14].

2.6.2. Calcium carbonates content

The carbonate content in the soil was measured by Collins Calcimeter as described in reference by (**Zhang and Bai, (2003)** [14].

2.6.3 Organic matter (OM) of soil

The Walkley and Black technique was used to calculate the total organic matter content [19] [20].

2.6.4 The soil peast extractable

It was prepared by taken 300 g from soil sample and added definite amount of distilled water until obtained on soil peast and the following determination methods were carried out, according to Jackson [21].

2.6.5 Electrical conductivity (EC)

Electrical conductivity (EC, ds/m) was measured in soil peast by means of conduct meter (cole parmer). The EC value gave us an indication about the total soluble salt [14].

2.6.6. Soluble cations and anions

The soluble cations and anions (me/L) were estimated in soil peast as followed [22]:

- Calcium and magnesium were estimated by titration with 0.01M versenate.
- Sodium and potassium were measured by using flame photometer as described in reference [23].
- Soluble Carbonate and bicarbonate were measured by titration with standard solution of 0.1 M hydrochloric acid; chlorides were measured by titration with 0.01M silver nitrate.
- Sulphates were measured by using the turbid metric method and measured by spectrophotometer at 440 nm [14].

2.7. Experimental layout

A pot experiment was carried out during October to January at the season of 2022 and 2023 in the experimental Shandawell station of Agriculture Research Centre, Sohag Governorate, Egypt to investigate the effect of treated wastewater on soil and plant properties. Three types of water have been applied to pots; treated with modified Bentonite and Bentonite- HA complex, treated with Bentonite and untreated wastewater from stock treated water during planting. After 15 days, it's were thinned to 5 seedlings. Plants were irrigated using the same volume of water in each time during the experiment. Chemical fertilizer (Urea and Potassium Phosphate) were added two as recommendation of Ministry of Agriculture.

3. Results and discussion

3.1. Characteristics of HA, Bentonite and Bentonite- HA complex.

3.1.1. XRD analysis

XRD patterns of the Bentonite are shown in Fig. (2); the figure was matched with pure bentonite XRD patter as represented elsewhere [24] .

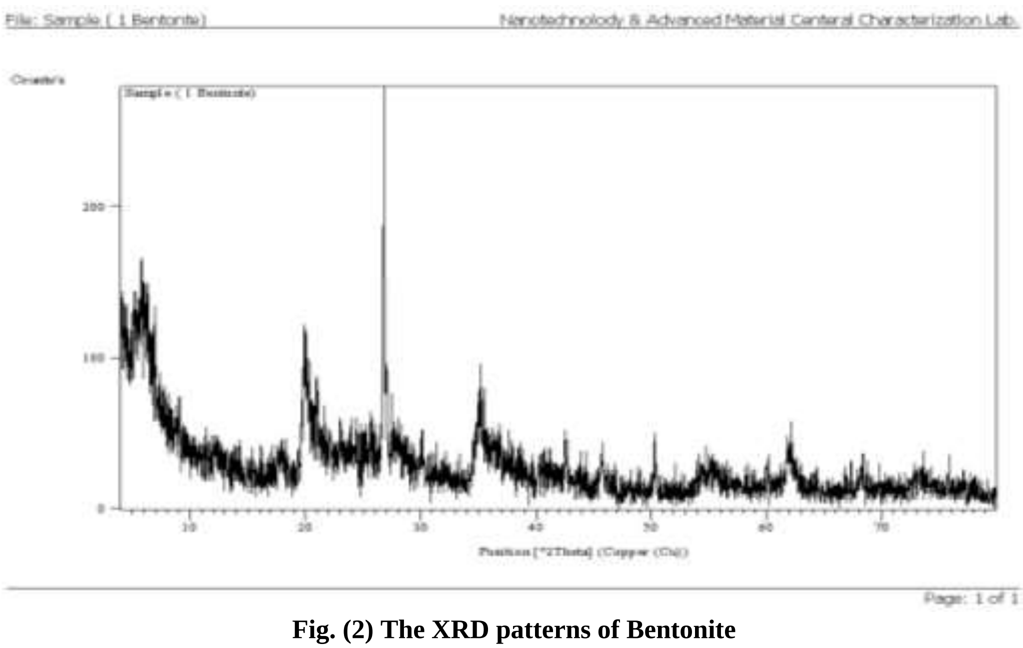

Fig. (2) The XRD patterns of Bentonite

3.1.2. Elemental analysis

The percentages of carbon, hydrogen, nitrogen, sulfur, and oxygen were 40.06, 4.57, 0.79, 0.73, and 53.83, respectively, according to the elemental analysis results for the Aldrich HA.

In addition, the chemical composition of Bentonite was determined by Energy Disperse Analysis X-ray (EDAX). The results showed that the chemical composition analysis of Bentonite (C 10.61%, O 48.92%, Mg 1.87%, Al 10.02 %, Si 22.73 %, K 2.08 %, and Fe 3.77%).

Figure (3) shows the Bentonite EDAX chart. According to Bentonite's elemental analysis, the structure is made up of a tetrahedral silicon sheet layered on top of an octahedral aluminum layer, with a Si: Al ratio of around 2:1.

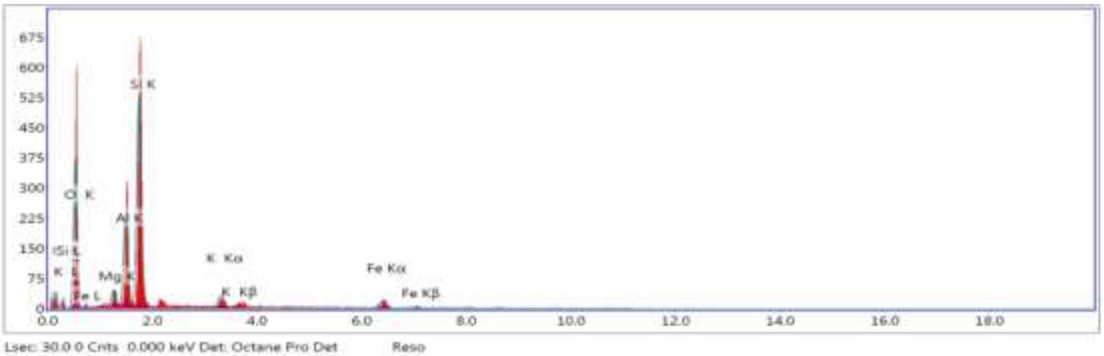

Fig. (3) Elemental analysis by EDAX

The changes in morphology following HA adsorption were examined using scanning electron microscopy (SEM). Bentonite and the Bentonite–HA complex are observed in SEM in Fig. (4), where a large number of tiny, distinct particles are dispersed throughout the microaggregate. The Bentonite-HA compound was discovered to have a rough surface. According to these findings, HA does aggregate Bentonite particles. In order to investigate the influence of HA on the surface, BET surface area for Bentonite and Bentonite-HA complex was determined.

BET surface of Bentonite and Bentonite–HA complex were 49.92 m^2g^{-1} and 59.4 m^2g^{-1}, respectively. Therefore, HA enhanced the surface and increased the active site of Bentonite.

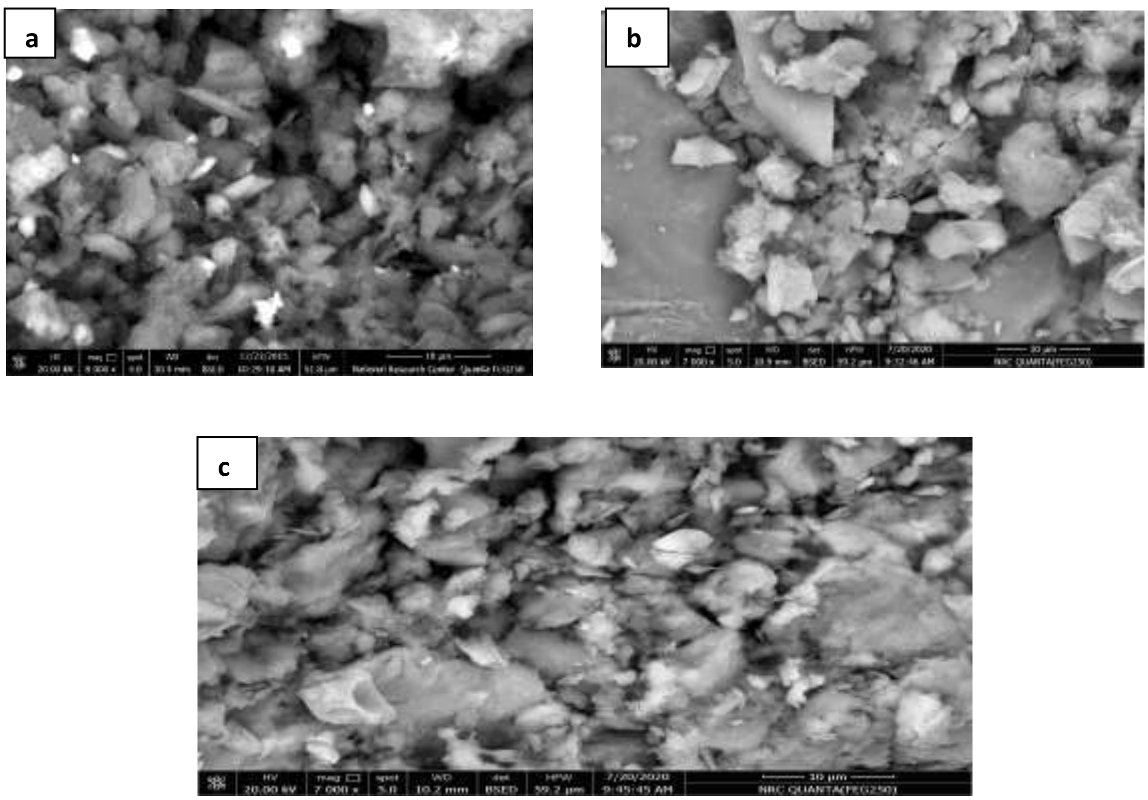

Fig. (4) SEM of a, b) Bentonite, C) Bentonite-HA complex

The conduct metric technique [23] was used as the second approach to assess the impact of HA on the Bentonite surface, total active site for Bentonite, and Bentonite – HA complex. Clearly, there were 5.71 meq g^{-1} and 6.7 meq g^{-1} of total active sites on Bentonite on the Bentonite–HA complex, respectively.

This indicates that under the same circumstances, HA has a larger functional group (COOH, -C=O, and -OH) than Bentonite. Additionally, the BET surface area has matched the results of the total active sites on Bentonite-HA complexes.

3.1.3. Zeta potential of Bentonite and Bentonite - HA complex

The result of zeta potential of bentonite and bentonite – HA complex was -25, and -27 (mV) respectively as shown in figure 5 and 6. The zeta potential clarified the ability of bentonite and bentonite – HA complex to adsorb heavy metals and major cations from wastewater, and the bentonite – HA complex has more capacity than bentonite in heavy metals and cations adsorption [25].

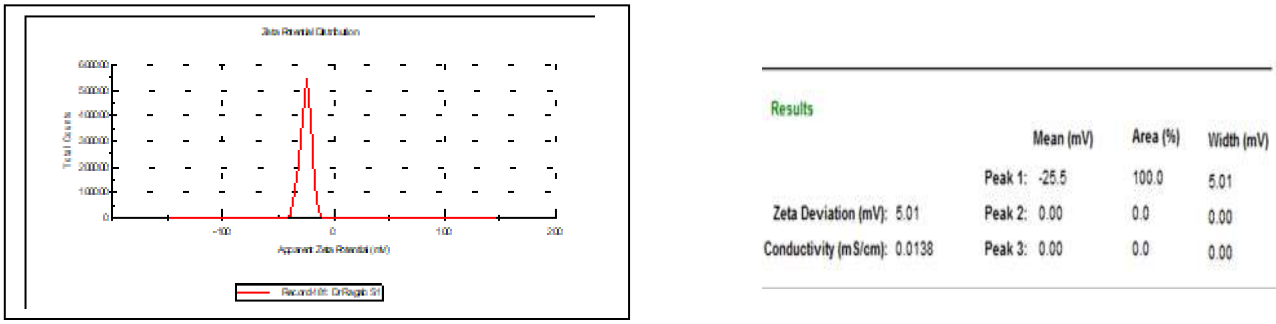

Fig. (5) Zeta potential distribution of Bentonite

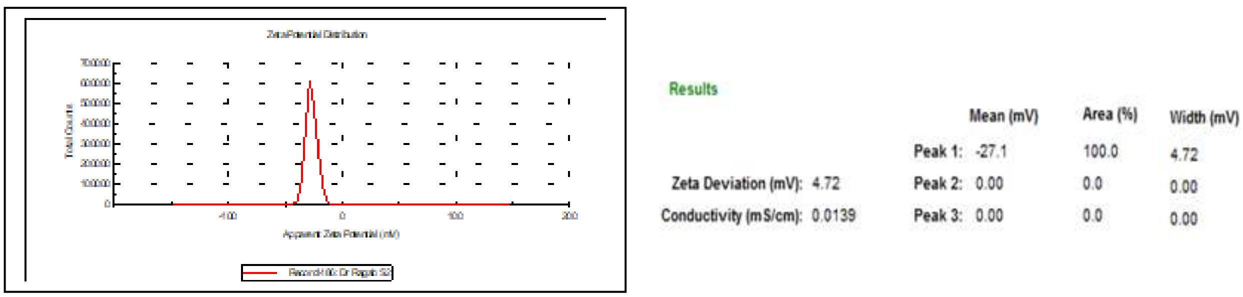

Fig. (6) Zeta potential distribution of Bentonite-HA complex

3.2. Wastewater treatment

Using a fixed bed column model, wastewater is treated by adsorption onto two sorbents: bentonite and bentonite-HA complex. For every 10 gm of Bentonite or Bentonite–HA complex, the rate flow was 200 and 250 ml/H, respectively.

The findings in Table 1 made it clear that when it applies to treating wastewater that contains organic contaminants and heavy metals, the Bentonite–HA complex performs better than Bentonite. This relates to the Bentonite-HA complex's higher BET surface area and highly active site contents compared to Bentonite. Additionally, HA often has a large number of functional groups (−COOH, −C=O, and −OH) attached to the aliphatic or aromatic carbons in the macromolecules, along with both hydrophobic and hydrophilic moieties. HA can therefore react with both organic and inorganic contaminants. The hydrophobic portion of HA then uses a hydrophobic-hydrophobic interaction mechanism to engage with organic contaminants, whereas carboxylic and hydroxyl groups use complexation and electrostatic processes to interact with heavy metals [26].

Table (1) chemical analysis of heavy metals and organic matter of wastewater)

Sample	Cd^{2+} ppm	Fe^{2+} ppm	Zn^{2+} ppm	Cu^{2+} ppm	Mn^{2+} Ppm	Co^{+} Ppm	Cr^{6+} ppm	Se^{4+} Ppm	Pb^{2+} ppm	TOC
Untreated WW	0.1	0.121	0.011	0.2	0.066	0.2	0.029	4.050	0.015	0. 6
Treated WW with Bentonite	0.9	0.119	0.010	0.11	0.050	0.095	0.0025	2.40	0.010	0.02
Treated WW with Bentonite -HA complex	0.79	0.000	0.000	0.07	0.031	0.002	0.019	2.02	0.005	0.00

By the same way, Bentonite and Bentonite – HA complex enhanced chemical analysis (EC, pH, major cations and major anions) of wastewater than Bentonite. The EC and pH values became much better to use in irrigation process. In addition, the ratio of cations and anions highly effected with treatment by Bentonite – HA complex than Bentonite. Wheresoever's, HA act as cheating agent.

Table (2) chemical analysis of cations and anions of wastewater

Sample no	pH	EC ds/m	Na^{+} mg/L	K^{+} mg/L	Ca^{2+} mg/L	Mg^{2+} mg/L	Cl^{-} mg/L	SO_4^{2-} mg/L	NO_3^{-} mg/L	CO_3^{-}	HCO_3^{-} mg/L
Untreated WW	7.75	1.97	9.36	0.69	5.62	3.45	4.06	8.96	23.54		6.12
Treated WW with Bentonite	7.11	1.56	4.69	0.65	4.50	2.24	3.38	4.46	17.22	-	4.24
Treated WW with Bentonite - HA complex	7.00	1.25	2.31	0.34	3.37	1.03	1.68	3.40	14.31	-	1.88

3.2.1. Effect of applied treated and untreated wastewater in irrigation

In order to evaluate the influence of irrigation with treated and untreated wastewater on some plant and soil properties under land experiment. Faba bean seeds were sown in soil under different irrigation with untreated and treated wastewater by Bentonite and Bentonite – humic complex in three replicated.

3.2.2. Effect of irrigation with treated and untreated wastewater on Faba bean properties

The characteristics of faba beans were listed in Table (3), including leaf area (cm2), stem diameter, plant height (cm), number of leaves per plant, fresh weight (g), dry weight (g), number of branches per plant, and fresh and dry weight of leaves under treated and untreated WW. The findings indicated that while the number of leaves, branches, leaf breadth, root length, and leaf area of the plot were not substantially impacted by the various water treatments, plant height, relative water content, chlorophyll a, chlorophyll b, and carotenoids parameters were.[26]

Table (3) Chemical Analysis for soil samples under study (in the peast extractable)

Treatment	Plant height (cm)	Relative water content%	Chlorophyll a (µg/ml)	Chlorophyll b(µg/ml)	Carotenoids(µg/ml)	dry weight (g)	fresh weight (g)	No of leaves	No of Branches	Leaf width (cm)	Root length (cm)	Average leaf area of pot (cm)
Untreated WW	14.00	74.6	15.99	27.78	4.90	0.0670	0.50	7.00	4.0	1.9	4.65	143.45
Treated WW with Bentonite	17.67	83.4	16.77	31.05	5.56	0.0787	0.657	9.00	5.3	3.0	4.22	165.14
treated WW with Bentonite – HA complex	22.50	82.2	17	30.15	5.08	0.084	0.689	10.00	6.1	4.1	5.57	177.89

3.3. Microbial test

Agriculture wastewater was subjected to pathogen analysis prior to treatment. One form of fungus known as Aspergillas sp. and two types of bacteria—both positive and negative from the Bacilli species were identified by pathogen analysis results.

To do this, wastewater was treated before chloride drops were applied. In the case of Bentonite and Bentonite-HA complex, the findings of a second pathogen study conducted after treatment demonstrated that the treated wastewater was devoid of bacteria and fungus.

4. Conclusion:

In conclusion, the present investigation demonstrated the high validity of removing heavy metals and organic contaminants in wastewater using with Bentonite -HA complex. The treatment process by studied materials will reduce environmental contamination. According to this study, the amounts of heavy metals, major cations, anions, and OM from WW were removed and reduced more effectively by the Bentonite-HA complex than by Bentonite alone. Additionally, the findings clarified how HA acid affects enhanced WW characteristics. According to the study's findings, TOC was entirely eliminated from WW.

Nonetheless, the TWW by Bentonite's heavy metals, SAR, TDS, EC, and pH were all within the permissible limit for irrigation. Additionally, test findings demonstrated that the chemical analysis was significantly impacted by the Bentonite and Bentonite-HA complex treatment method.

Bentonite -HA complex had a significant effect on some chemical analysis than the treatment by Bentonite. Hence, Bentonite and Bentonite -HA complex offers an efficient and low-cost approach for wastewater treatment and using it for irrigation purposes.

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