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Integrated treatment of black wastewater using sedimentation, constructed wetland, and nanoparticles coagulation

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ABSTRACT

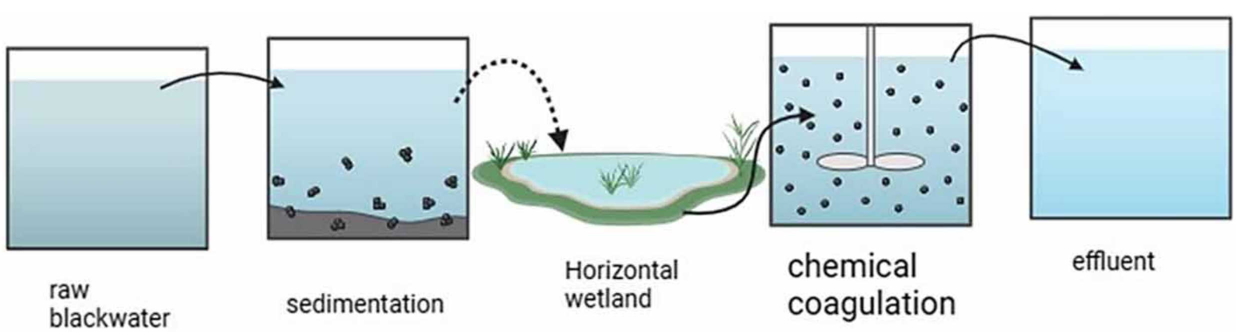
The aim of the present investigation was to achieve an efficient treatment for black wastewater for non-restricted reuse. The treatment included sedimentation followed by horizontal flow constructed wetland (HF-CW), and finally chemical coagulation using magnetite (Fe_3O_4) nanoparticles. For the wetland, the average organic loading rate (OLR) was 42.83 biochemical oxygen demand (BOD)/m^2 day and 71.30 g chemical oxygen demand (COD)/m^2 day, while average surface loading rate was 0.185 m^3/m^2 day. The obtained results revealed that removal percentage of COD, BOD, and total suspended solids (TSS) by sedimentation followed by processing through wetlands was 80.3, 81.6, and 80%, which corresponds to 76, 42.56, and 54 mg/l, respectively. When magnetite nanoparticles were added to the HF-CW effluent at the optimum dose of 25 mg/l in combination with 10 mg/l $FeCl_3$, the overall removal rate increased to reach 95.9, 99.1, 99.17, 92.3, 94.3, 94.3, and 91.3% for turbidity, COD, BOD, TSS, phosphate (PO_4), total nitrogen (TN), and sulfide, respectively. The corresponding residual concentrations were 5 NTU, 8, 5, 18 according to table 3, 0.07, 1, and 1.04 mg/l, respectively. According to the national and international regulations, the present final treated effluent can be safely reused as a non-restricted source for agriculture purposes. The overall results revealed that the implemented study is simple, efficient, reliable, and economical, particularly for remote areas, according to the non-restricted reuse requirements.

Key words: blackwater treatment, chemical coagulation, constructed wetland, nanoparticles, non-restricted water reuse, sedimentation

HIGHLIGHTS

- Integration of sedimentation/wetland processes is efficient for treatment of blackwater.
- The removal of pollutants via sedimentation/wetland complies with national regulations.
- Chemical coagulation using nanoparticles is efficient for treatment of blackwater.
- The overall removal of pollutants' treatment train complies with the non-restricted reuse regulation.

GRAPHICAL ABSTRACT



1. INTRODUCTION

Approximately two-thirds of our Earth's surface is, certainly, covered by water, making it one of the most plentiful natural resources. However, many nations around the globe, particularly those countries in the Middle East and North Africa (MENA) are facing a serious lack of access to clean and/or safe drinking water (Emenike *et al.* 2017). Water scarcity has been identified as the greatest global threat facing humanity (Deshpande *et al.* 2020). In addition, water contamination is now a major global concern (Jhansi & Mishra 2013). Conversely, world population growth, industrialization, and inadequate wastewater treatment (WWT) are all to be blamed for the global concern (Chowdhary *et al.* 2020; Maurya *et al.* 2020; Zhongming *et al.* 2020). The primary sources of pollution in water resources worldwide are the discharge of different wastewaters (WWs) including industrial activities, municipal effluents, hospital wastes, and/or urban stormwater runoff (Wang *et al.* 2014; Zhongming *et al.* 2020). Therefore, immediate action should be taken to manage such a problem correctly. In this respect, it has been predicted that the quality of diverse water resources in the MENA countries will continue to worsen as the most threatening problem (Zhongming *et al.* 2020). Due to the global rise in water scarcity today, effective WWT and safe reuse through a circular economy (CE) are urgently needed (Jhansi & Mishra 2013).

Conversely, black WW is generally composed of flushing water, faecal matter, and urine. It accounts for 27–30% of domestic sewage. Furthermore, grey WW accounts for 70–73% of such domestic sewage. The latter originates from the daily showers, washing basins, laundry, and so on. (Zhou *et al.* 2020). Black WW is a major potential source of pollution in the developing countries due to lack of sanitation facilities. The direct discharge of untreated black WW can lead to the contamination of the environment including soil and water ways. This in turn creates problems with drinking water and food. Thus, high-efficiency and economical technologies are urgently needed for the effective treatment of such blackwater to reduce environmental pollution and avert the impending water crisis (Xu *et al.* 2023). Therefore, appropriate WWT and safe recycling are strongly required owing to the rising demand for clean, inexpensive water worldwide (Kumar & Pal 2018; Leader & Wijnen 2018). When WW is properly handled, it can be safely reused for different purposes according to the degree of treatment. To solve the water crisis, countries have adopted rules and regulations for WW assessment and remediation (Abdel-Shafy *et al.* 2017; Miarov *et al.* 2020).

Nevertheless, adequate WWT and/or upgrading/upscaling are all great challenges and costly due to their capital expense. It is a reliable alternative system to sewage treatment (Abdel-Shafy & Dewedar 2012; Abdel-Shafy & Mansour 2018; Abdel-Shafy & Mansour 2019; Abdel-Shafy *et al.* 2022a, b). In this context, it has been confirmed that constructed wetlands (CWs) have been successfully employed for the treatment of several domestic WW sources. The CWs have proved to be simple, and low-cost in construction, operation, maintenance, and management (Abdel-Shafy *et al.* 2017, 2022a, b, 2023; El-Khateeb & Abdel-Shafy 2022). Consequently, the application of CWs for WWT has been recently proposed as a reliable alternative to other techniques and water reuse measures (Abdel-Shafy & Mansour 2020; Mainardis *et al.* 2022).

It was reported that black WW was treated efficiently in Egypt by employing sedimentation, followed by horizontal then vertical wetlands treatment (Abdel-Shafy *et al.* 2022a, b). However, the final treated effluent could only cope as a secondary treated effluent.

Conversely, nanoparticles (NPs) have been extensively applied in environmental remediation due to their high surface area and efficiency in WWT (Li *et al.* 2022). An appropriate amount of NPs in WWT proved to upgrade the quality of the final treated effluent from 69 to 92% according to different researchers (Fouda *et al.* 2021; Khilji *et al.* 2022).

The present study aimed to evaluate the efficiency of combining the sedimentation process, CWs, and subsequent chemical coagulation using NPs for the treatment of blackwater. The final treated effluent was evaluated for the purpose of safe reuse.

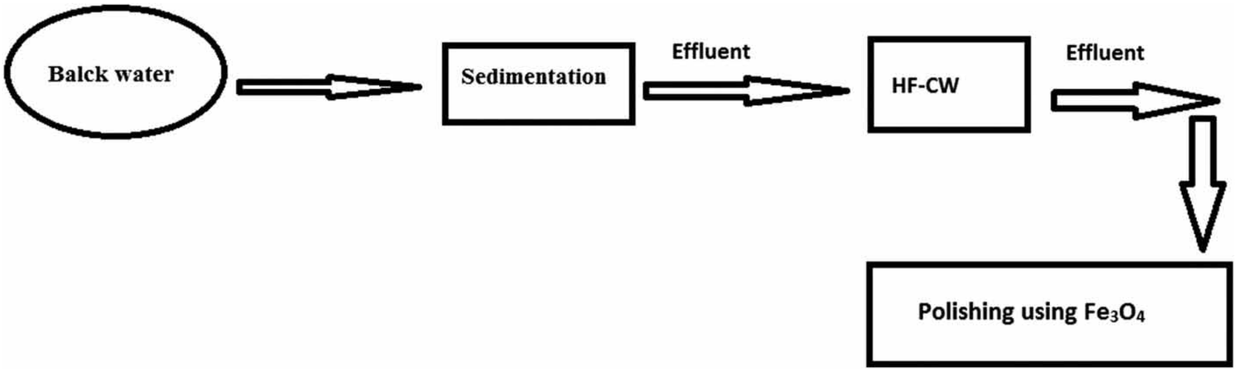
2. MATERIALS AND METHODS

2.1. Source of raw black WW

The source of black wastewater was obtained by a piping separation system from a house across the street to a pilot plant area in the National Research Centre (NRC), Cairo, Egypt. The piping systems separate black, grey, and yellow WW. Each type of piping system was connected to a separate manhole. The black and grey waters were raised separately to the sedimentation tanks (STs) using electric pumps. The STs were erected at a height of 4 m each.

2.2. Pilot plant for black WWT

The following diagram presents the train of the studied black wastewater treatment system.



2.2.1. Sedimentation tanks

The blackwater STs are shown in Figure 1. The latter consists of three successive STs as a primary treatment system. The dimensions of these STs are 1.2 × 1.2 × 1.2 m each, and the wastewater flow rate is 1,500 L/day (Figure 1).

2.2.2. Horizontal flow constructed wetland (HF-CW) for black WWT

The outlet from the ST was directed to the horizontal flow constructed wetland (HF-CW) (Figure 2).

The dimensions of the HF-CW are 3.0 × 1.5 × 0.9 m (Supplementary Table S1). In addition, Supplementary Table S1 presents the operating conditions of the investigated HF-CW, including the wastewater flow rate, organic loading rate (OLR), surface loading rate (SLR), and the hydraulic retention time (HRT).

2.2.3. Chemical coagulation using different doses of synthesized magnetite NPs (Fe_3O_4)

2.2.3.1. Preparation of magnetite NPs (Fe_3O_4). **Materials:** Ferric nitrate [Fe $(NO_3)_3 \cdot 9H_2O$] (from Qualikems India) and ethylene glycol ($C_2H_6O_2$) (from Oxford Lab. Chem, purchased from Mumbai India) – analytical grade (the purity is more than 95%) were used to prepare the magnetite ferric oxide NPs (Fe_3O_4).

Synthesis of magnetite NPs (Fe_3O_4): The procedure of synthesizing magnetite NPs is described as follows:

Prepare 0.2 mol ferric nitrates dissolve in 100 ml ethylene glycol followed by vigorous stirring for 2 h at 40 °C. The obtained solution is heated to 80 °C and kept at this temperature until obtaining a brown gel. The obtained gel is kept at room temperature for about 1 h. The obtained gel is annealed at 200 °C in a furnace under an air atmosphere. Finally, the magnetite NPs are synthesized as (Fe_3O_4), according to Jing Xu *et al.* (2007).

HRTEM (JEOL JEM-2100) electron microscope is used to gain the actual particle size of the synthesized Fe_3O_4. This particle size was found to be less than 50 nm, as shown in Figure 3.

X-ray diffraction (XRD; PANalytical Empyrean) is employed under the following conditions: scan axis: Gonio, start position $2\theta = 5.0129°$, end position $2\theta = 79.9709°$, step size $2\theta = 0.0260°$, scan step time at 18.8700 Sec, scan type: continuous,

Figure 1 | Three successive STs as a primary treatment of black WW.

Figure 2 | HF-CW pilot plant.

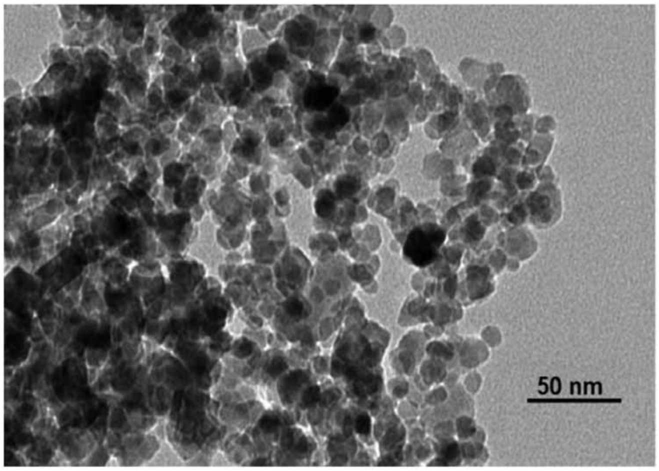


Figure 3 | Particle size of the prepared Fe_3O_4 NPs.

PSD length $2\theta = 3.35°$, measurement temperature at 25 °C, anode material: Cu-Kα1 at 1.54060 Å, Cu-Kα2 at 1.54443 Å, Cu-Kβ at 1.39225 Å, ratio Kα2/Kα1 = 0.50000, generator settings: 30 mA, 45 kV.

Figure 4 presents the XRD pattern of the synthesized Fe_3O_4 NPs, as well as the purity of the synthesized NPs.

Scanning electron microscope (SEM; JEOL-JSM-5400, Japan) with the magnification of 30 kV ×1,000 was employed in the present study to measure the surface morphology. Figure 5 exhibits the surface morphology of the synthesized Fe_3O_4 NPs.

2.3. Bench-scale chemical coagulation using different doses of the synthesized Fe_3O_4 NPs

The jar test procedure as a bench-scale experiment was employed according to the procedure reported by Abdel-Shafy (2015). The effluent of HF-CW was subjected to different doses of the synthesized Fe_3O_4 NPs, as follows: 5, 10, 15, 20, and 25 mg/l, in five different individual jars containing 1 L each of the HF-CW effluent. All the physicochemical characteristics of the black

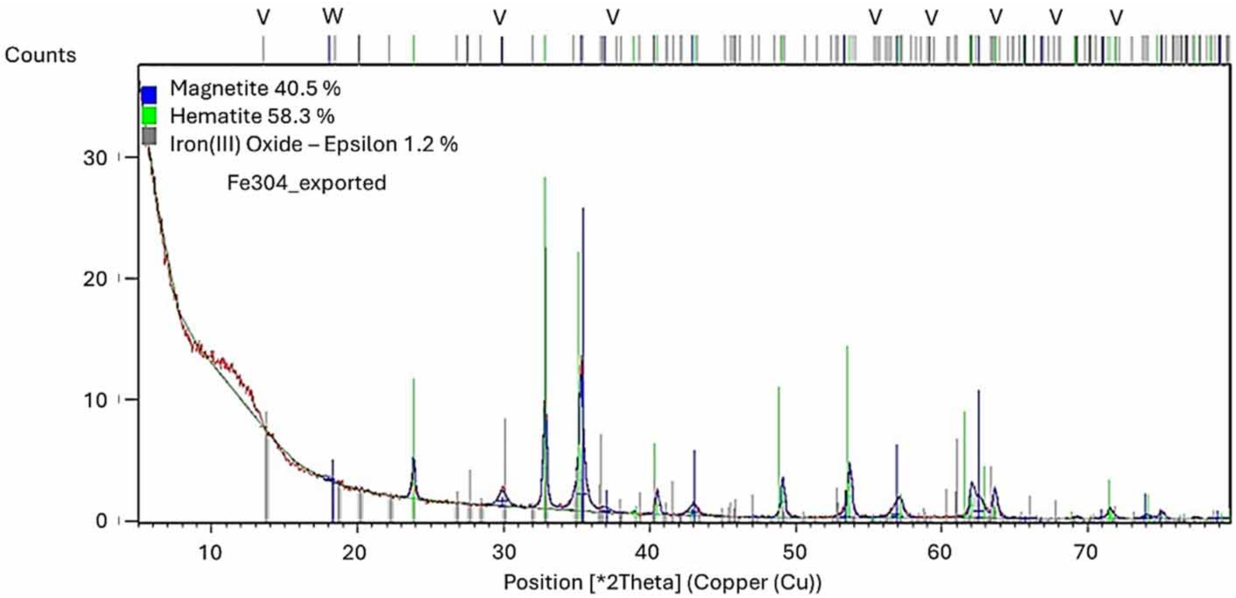


Figure 4 | XRD pattern of Fe_3O_4.

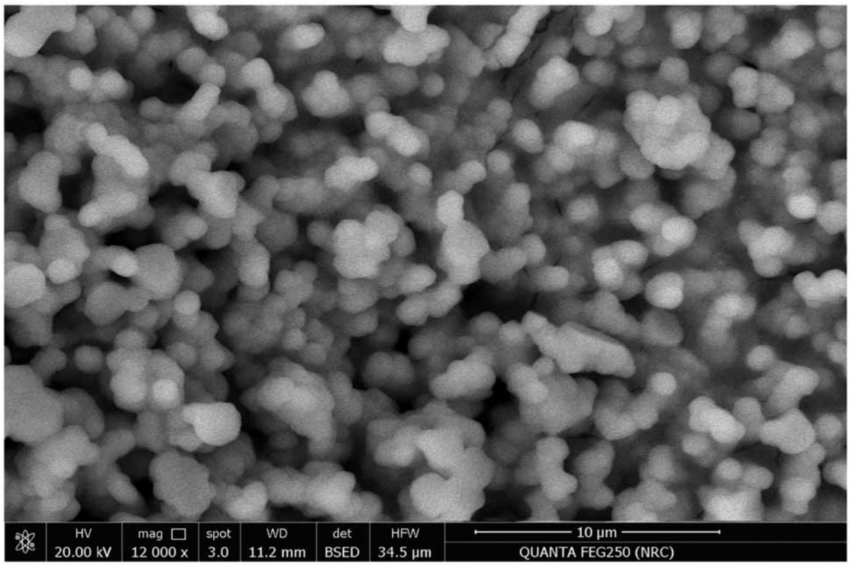


Figure 5 | SEM image as the surface morphology of the synthesized Fe_3O_4 NPs.

WW throughout this study were determined according to the Standard Methods (Baird 2017). Therefore, the optimum dose of Fe_3O_4 NPs was determined. Each experiment was conducted six times. The given results are the average of all the studied experiments. The standard deviation of each result was calculated accordingly.

3. RESULTS AND DISCUSSION

3.1. Raw black WW

Supplementary Table S2 presents the physical/chemical characteristics of the raw black WW. The levels of COD, BOD, and TSS were 890, 605, and 415 mg/l, respectively. This black WW is considered to be within the category of medium-strength WW.

3.2. ST

The raw black WW was subjected to the sedimentation process as a primary treatment. The retention time (RT) was 24 h, as indicated in Supplementary Table S2. Figure 6 presents the performance of ST for the treatment of black WW. The effluent of the ST indicated that the levels of COD, BOD, and TSS were reduced from 890, 605, and 415 mg/l to 386, 232, and 270 mg/l, respectively, (i.e., at a corresponding removal rate of 56.6, 61.7, and 34.9%, successively as shown in Supplementary Table S2. In addition, the removal of phosphates, total Kjeldahl nitrogen (TKN), and sulfides were achieved at the rate of 54.0, 43.0, and 41.7%, respectively. The BOD/COD ratio decreased from 0.68 to 0.60 indicating a slight improvement in the biological degradation of the primary treated black WW (Rajagopal *et al.* 2019; Abdel-Shafy *et al.* 2023). The main mechanisms for TSS removal using the ST are hydrolysis, sedimentation, and biotransformation processes during the biological treatment (Abdel-Shafy *et al.* 2017; Rajagopal *et al.* 2019; Zehra *et al.* 2019).

The efficiency of the ST for the removal of certain pollution parameters is shown in Figure 6.

3.3. Directing the ST effluent to HF-CW

The effluent of ST was directed to the HF-CW at a SLR of 0.185 m^3/m^2 day and an average organic loading rate (OLR_{Avg}) of 42.83 g BOD/m^2 day and 71.30 g COD/m^2day. Supplementary Table S3 presents the efficiency of the HF-CW for the treatment of ST effluent. The performance of the HF-CW for the removal of COD, BOD, and TSS was 80.3, 81.6, and 80%, respectively (Figure 7).

Different mechanisms supported the performance of HF-CW. These mechanisms include filtration, sedimentation, plant uptake, and biotransformation, respectively. Further reduction of the BOD/COD ratio indicates the biodegradation of the organic loads (Abdel-Shafy & El-Khateeb 2013; Abdel-Shafy *et al.* 2023).

3.3.1. Bench-scale chemical coagulation using Fe_3O_4 NPs

Chemical coagulation of the HF-CW effluent using Fe_3O_4 NPs at different doses was conducted using the bench-scale jar test apparatus. The purpose is to study the efficiency of Fe_3O_4 NPs for the treatment of CW effluents in terms of removing the pollution parameters. The results are provided in Table 1. These results indicate that increasing the Fe_3O_4 NP dose increases the treatment efficiency. The highest removal rate was obtained by using 25 mg/l Fe_3O_4 NPs. However, it was observed that the iron oxide NPs produced fine flocs that were suspended for more than 30 min. Therefore, it was important to enhance the precipitation of these flocs.

Coagulation is a process of combining small particles into larger and relatively heavier aggregates (flocs). Then precipitates the particulate aggregates consequently these impurities can be removed in subsequent solid/liquid separation processes. The smaller nanoparticles are more efficient than larger particles in terms of coagulation process (Jiang 2015).

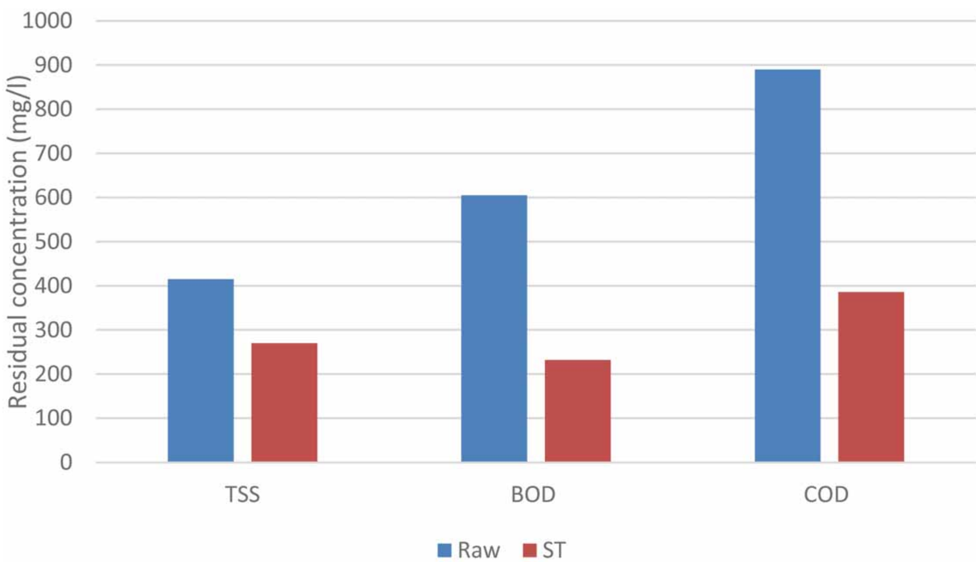


Figure 6 | Levels of COD, BOD, and TSS in the influent and effluent of black WW before and after sedimentation.

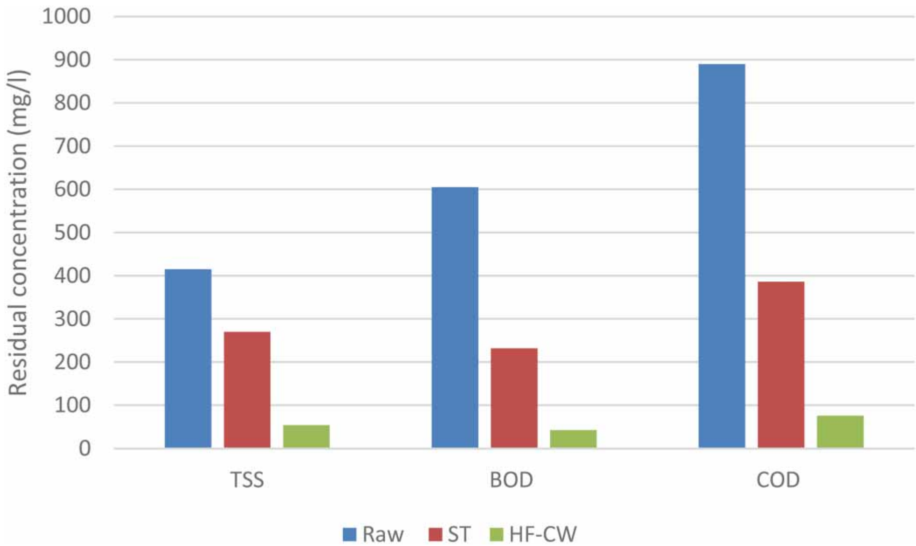


Figure 7 | Levels of BOD, COD, and TSS in the raw black wastewater (a) after sedimentation and (b) after processing in the CW.

3.3.2. Bench-scale chemical coagulation using Fe_3O_4 NPs and $FeCl_3$ as coagulant aid

For the purpose of enhancing the settling of the coagulated particles, it was important to add $FeCl_3$, which is well known as an efficient coagulant aid. By adding 10 mg/l $FeCl_3$ in combination with variable doses of Fe_3O_4 NPs, as presented in Table 2, a notable settling efficiency was recorded. This could be attributed to the fact that $FeCl_3$ enhances the precipitation of the fine flocs by gathering them together to form larger and heavier ones. Hence, relatively heavier flocs precipitate more efficiently (Liu *et al.* 2021). Consequently, better removal of TSS was obtained (Table 3). The results (Table 3) reveal that the optimum dose is 25 mg/l Fe_3O_4 NPs in combination with 10 mg/l $FeCl_3$. The recorded removal efficiency of turbidity, COD, and BOD was 64.3, 89.5, and 88.3%, respectively. The residual concentrations of turbidity, COD, and BOD in the effluent were 5 NTU, 8, and 5 mg/l, respectively (Table 3).

3.3.2.1. Discussion and conclusions. The present study deals with black WW characterized by medium strength according to. The ratio of BOD/COD was 0.68 indicating the biological degradation. By implementing the three successive processes, sedimentation via tanks at 24 h retention time could eliminate the pollution parameters of COD, BOD, TSS, phosphates, TKN, and sulfides at the rate of 56.6, 61.7, 34.9, 54.8, 43.8, and 41.7, respectively. The BOD/COD ratio decrease from 0.68 to 0.60 indicated the biodegradation of the given organic load (Regelsberger *et al.* 2007; Masi *et al.* 2010; Capodaglio 2017).

The sedimentation effluent was further treated by the horizontal wetland system at an OLR of 42.83 g BOD/m^2 day and 71.30 COD/m^2 day. The obtained removal rates of the COD, BOD, and TSS reached 80.3, 81.6, and 80%, respectively. By employing the successive treatment systems, the studied pollution parameters could be successfully eliminated. Such elimination by use of wetlands could be attributed to several factors including RT, nutrients uptake by plants, and biomass formation (Xu *et al.* 2007; Abdel-Shafy *et al.* 2017). The BOD/COD ratio indicated the biodegradation of the organic load in the given treated wastewater.

By employing chemical coagulation to the effluent of CW using different doses of the Fe_3O_4 NPs, the characteristics of the treated effluent improved and the optimum dose proved to be 25 mg/l. However, removal of the suspended solids was not satisfactory. Hence, by adding 10 mg/l of $FeCl_3$ in combination with 25 mg/l of the Fe_3O_4 NPs, the final treated wastewater was greatly improved. Consequently, no release of any non-dissolved nanoparticles was recorded. This can be attributed to the use of $FeCl_3$ in terms of settling of the coagulated particles (Jagaba *et al.* 2018; Deng & Abdel-Shafy 2024).

It was reported that alum and $FeCl_3$ were employed separately for the treatment of WW (Jagaba *et al.* 2018; Ettalouia *et al.* 2021). However, much larger doses of these coagulants were used. Therefore, by using small doses of the employed NPs and coagulant two advantages were achieved, namely, efficiency and cost-effectiveness.

Table 1 | Characteristics of HF-CW effluent before and after addition of different doses of Fe_3O_4 nanoparticles

			Dose of Fe_3O_4 (nanoparticles)									
			5 (mg/l)		10 (mg/l)		15 (mg/l)		20 (mg/l)		25 (mg/l)	
Parameter	**Repeatability of experiments**	**HF-CW effluent**	**Conc. ± STDE**	**% R**	**Conc. ± STDE**	**% R**	**Conc. ± STDE**	**% R**	**Conc. ± STDE**	**% R**	**Conc. ± STDE**	**% R**
pH	6	8.62 ± 0.43	8.47 ± 0.42	–	8.3 ± 0.42	–	8.22 ± 0.4	–	8.16 ± 0.041	–	8.15 ± 0.40	–
EC (mS/Cm)	6	1,740 ± 87	1,566 ± 78.3	10	1,305 ± 65.25	25	1,218 ± 61	30	1,131 ± 57	35	870 ± 44	50
TDS (mg/l)	6	1,043 ± 52	834.4 ± 41.7	20	782.25 ± 40	25	730 ± 37	30	625.8 ± 30	40	573.65 ± 27	45
Turbidity (NTU)	6	14 ± 0.7	17 ± 0.85	–	16 ± 0.8	–	15 ± 0.75	–	18 ± 0.9	–	17 ± 0.85	—
COD (mg/l)	6	76 ± 3.8	60 ± 3	21.0	40 ± 2	47.4	30 ± 1.5	60.5	20 ± 1.1	73.7	10 ± 0.51	86.8
BOD (mg/l)	6	42.56 ± 2.1	38.5 ± 1.93	9.5	30.0 ± 1.5	29.5	20.05 ± 1.0	52.9	12.88 ± 0.64	69.7	9.1 ± 0.46	78.6
TSS (mg/l)	6	54 ± 2.7	51 ± 2.55	5.5	52 ± 2.6	3.7	53 ± 2.65	1.85	55 ± 2.75	–	56 ± 2.8	–
PO_4 (mg/l)	6	0.09 ± 0.005	0.088 ± 0.004	2.2	0.085 ± 0.004	5.5	0.08 ± 0.004	11.1	0.078 ± 0.08	13.3	0.075 ± 0.004	16.6
TN (mg/l)	6	1.87 ± 0.09	1.5 ± 0.075	19.7	1.35 ± 0.07	27.8	1.25 ± 0.06	33.2	1.24 ± 0.06	33.7	1.21 ± 0.02	35.3
Sulfide (mg/l)	6	1.34 ± 0.07	1.32 ± 0.066	1.49	1.29 ± 0.06	3.7	1.27 ± 0.06	5.2	1.23 ± 0.06	8.2	1.2 ± 0.06	10.4

Table 2 | Characteristics of HF-CW effluent before and after addition of different doses of Fe_3O_4 nanoparticles in combination with 10 mg/l of $FeCl_3$

			Different doses of Fe_3O_4 with 10 mg/l $FeCl_3$									
			5 (mg/l) Fe_3O_4		10 (mg/l) Fe_3O_4		15 (mg/l) Fe_3O_4		20 (mg/l) Fe_3O_4		25 (mg/l) Fe_3O_4	
Parameter	***N***	**HF-CW effluent**	**Conc. ± STDE**	**% R**	**Conc. ± STDE**	**% R**	**Conc. ± STDE**	**% R**	**Conc. ± STDE**	**% R**	**Conc. ± STDE**	**% R**
pH	6	8.62 ± 0.4	7.35 ± 0.37	–	7.31 ± 0.4	–	7.28 ± 0.43	–	7.23 ± 0.36	–	7.21 ± 0.40	–
EC (mS/Cm)	6	1,740 ± 87	1,450 ± 73	16.6	1,200 ± 61	31	1,185 ± 60	32	890 ± 44.5	48.8	680 ± 34	61
TDS (mg/l)	6	1,043 ± 52.2	803.1 ± 40.2	23	730.1 ± 36.5	30	667.52 ± 34	36	532.1 ± 27	49	417.2 ± 21	60
Turbidity (NTU)	6	14 ± 0.71	12 ± 0.5	14.2	10 ± 0.4	28.6	9 ± 0.45	55.5	6 ± 0.36	57.1	5 ± 0.25	64.3
COD (mg/l)	6	76 ± 3.7	52 ± 2.5	31.6	32 ± 1.6	57.8	27 ± 1.6	64.5	17 ± 0.85	77.6	8 ± 0.48	89.5
BOD (mg/l)	6	42.56 ± 2.13	36 ± 1.7	15.4	21 ± 1.05	50.6	17 ± 1.02	60.05	11 ± 0.55	74.2	5 ± 0.3	88.3
TSS (mg/l)	6	54 ± 2.7	43 ± 2.2	20.4	37 ± 2.2	31.5	32 ± 1.6	40.7	24 ± 1.8	55.5	18 ± 1.6	66.7
PO_4 (mg/l)	6	0.09 ± 0.005	0.08 ± 0.004	11.1	0.078 ± 0.003	13.3	0.076 ± 0.004	15.5	0.074 ± 0.004	17.7	0.070 ± 0.003	22.2
TN (mg/l)	6	1.87 ± 0.1	1.46 ± 0.07	22	1.30 ± 0.065	30.48	1.15 ± 0.07	38.5	1.20 ± 0.06	35.8	1 ± 0.05	46.5
Sulfide (mg/l)	6	1.34 ± 0.07	1.3 ± 0.06	3	1.25 ± 0.064	6.7	1.20 ± 0.06	10.4	1.18 ± 0.07	12	1.04 ± 0.05	22.4

Table 3 | The overall percentage of removal using the combined treatment systems

Parameter	**Unit**	**N**	**Raw WW**	**ST**	**HF-CW**	**[a]25 mg/l Fe_3O_4 with 10 mg/l $FeCl_3$**	**Overall removal %R**
pH	–	6	8.65 ± 0.4	8.7 ± 0.5	8.62 ± 0.51	7.21 ± 0.36	–
EC	mS/Cm	6	2,340 ± 117	2,580 ± 129	1,740 ± 87	680 ± 34	71
TDS	mg/l	6	1,404 ± 70.2	1,543 ± 77.2	1,043 ± 52.15	417.2 ± 21	70.3
Turbidity	NTU	6	122 ± 6.1	70 ± 3.5	14 ± 0.7	5 ± 0.35	95.9
COD	mg/l	6	890 ± 44.5	386 ± 19.3	76 ± 3.8	8 ± 0.4	99.1
BOD	mg/l	6	605 ± 30.3	232 ± 14	42.56 ± 2.1	5 ± 0.3	99.17
BOD/COD	–	6	0.68 ± 0.04	0.6 ± 0.03	0.56 ± 0.033	0.62 ± 0.03	–
TSS	mg/l	6	415 ± 20.8	270 ± 16.2	54 ± 2.7	18 ± 1.6	95.7
PO_4 (total)	mg/l	6	2.13 ± 0.11	0.98 ± 0.05	0.09 ± 0.004	0.070 ± 0.004	96.7
TN	mg/l	6	64 ± 3.2	36 ± 2.2	1.87 ± 0.1	1 ± 0.05	94.3
Sulfide	mg/l	6	12 ± 0.72	7 ± 0.35	1.34 ± 0.07	1.04 ± 0.05	91.3
Number of cells or eggs of nematode	Count/l	3	–	–	–	ND	–

[a]After addition of Fe_3O_4 as chemical coagulant in combination with 10 mg/l $FeCl_3$ as coagulant aid.
Raw WW, raw black wastewater; ST, sedimentation tanks effluent; HF-CW, horizontal flow constructed wetlands effluent; N, experimental replicate.

By correlating the final overall treated effluent with the permissible limits of the national and international regulation for non-restricted reuse (Supplementary Table S7), it can be confirmed that the obtained final treated effluent can be safely reused for irrigation without any restriction.

In comparison of the present study with the previous work (Abdel-Shafy *et al.* 2017), where two types of wetlands were employed. Thus, larger area was necessary to be used, including the cost of labour and maintenance. Besides, the final treated effluent was classified as secondary treated effluent. By comparing the present results with the previous one (Abdel-Shafy *et al.* 2017), it was observed that the latter could achieve the following: (1) only one CW (horizontal) as land area (i.e. relatively smaller land is required), (2) less labour and maintenance cost, and (3) a tertiary treated effluent could be achieved, inside of secondary treated effluent. Hence, three important advantages were reached.

3.3.2.2. Recommendation. High-strength WW such as blackwater can be treated efficiently by employing the studied successive processes, namely, sedimentation, CWs, and subsequently chemical coagulation using Fe_3O_4 NPs in combination with small dose of $FeCl_3$. The final treated effluent can be safely reused in irrigation as a non-restricted source of water. Three different advantages were achieved by employing the present study.

ACKNOWLEDGEMENTS

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DATA AVAILABILITY STATEMENT

All relevant data are included in the paper or its Supplementary Information.

CONFLICT OF INTEREST

The authors declare there is no conflict.

REFERENCES

Abdel-Shafy, H. I. (2015) Chemical treatment for removal of heavy metals from industrial wastewater, *Egyptian Journal of Chemistry*, **58** (1), 1–12.

Abdel-Shafy, H. I. & Dewedar, A. (2012) Constructed wetlands for urban wastewater treatment in Egypt, *Sustainable Sanitation Practice*, **12**, 27–32.

Abdel-Shafy, H. I. & El-Khateeb, M. (2013) Integration of septic tank and constructed wetland for the treatment of wastewater in Egypt, *Desalination and Water Treatment*, **51** (16–18), 3539–3546.

Abdel-Shafy, H. I., Mansour, M. S. M. (2018) Phytoremediation for the elimination of metals, pesticides, PAHs, and other pollutants from wastewater and soil. In: Chandra, R., Gurjan, B. R. & Govil, J. N. (eds.) *Phytobiont and Ecosystem Restitution*, Vol. 6. Singapore: Springer Nature, pp. 101–136. *Environmental Science and Engineering*, Vol. 6, 31 December 2018. Toxicology, Publisher: STUDIUM PRESS LLC, USA.

Abdel-Shafy, H. I. & Mansour, M. S. M. (2019) *Microfiltration Overview: A Promising Technique for Wastewater Treatment and Reuse, Chapter 3*. In: Rayess, Y. E. (ed.) *Microfiltration: Principles, Process and Applications*, New York: NOVA SCIENCE PUBLISHER Inc, pp. 73–158.

Abdel-Shafy, H. I. & Mansour, M. S. M. (2020) Rehabilitation and upgrading wastewater treatment plant for safe irrigation reuse in remote area, *Journal of Water Practice and Technology*, **15** (4), 1213–1227.

Abdel-Shafy, H. I., El-Khateeb, M. A. & Shehata, M. (2017) Blackwater treatment via combination of sedimentation tank and hybrid wetlands for unrestricted reuse in Egypt, *Journal of Desalination and Water Treatment*, **71**, 145–151.

Abdel-Shafy, H. I., El-Khateeb, M. A., Ahmed, H. M., Hefny, M. M. & Abdel-Haleem, F. M. (2022a) Greywater treatment for safe recycling via hybrid constructed wetlands and sludge evaluation, *Egyptian Journal of Chemistry*, **65**, 543–555.

Abdel-Shafy, H. I., El-Khateeb, M. A. & Mansour, M. S. (2022b) Constructed wetlands for wastewater management in Egypt: An overview of 30-years experiences in small/medium-size treatment plants. In: *Constructed Wetlands for Wastewater Treatment in Hot and Arid Climates*, Springer, Cham, pp. 71–99. https://doi.org/10.1007/978-3-031-03600-2_5.

Abdel-Shafy, H. I., Ahmed, H. M., El -Khateeb, M., Sobhy, N. A., Hefny, M. M. & Abdel-Haleem, F. M. (2023) Constructed wetland enhancement via chemical coagulation using variable hydraulic retention times for grey wastewater treatment, *Egyptian Journal of Chemistry*, **66**, 55–67 (accepted for publication).

Baird, R. B. (2017) *Standard Methods for the Examination of Water and Wastewater*, 23rd edn. American Public Health Association (APHA). Water Environment Federation, Washington, DC.

Capodaglio, A. G. (2017) Integrated, decentralized wastewater management for resource recovery in rural and peri-urban areas, *Resources*, **6** (2), 22.

Chowdhary, P., Bharagava, R. N., Mishra, S. & Khan, N. (2020) Role of industries in water scarcity and its adverse effects on environment and human health, *Environmental Concerns and Sustainable Development: Air, Water and Energy Resources*, **1**, 235–256.

Deng, Y. & Abdel-Shafy, H. I. (2024) Barriers to ferrate(VI) application in water and wastewater treatment, *Environmental Science & Technology*, **58**, 3057–3060.

Deshpande, B., Agrawal, P., Yenkie, M. & Dhoble, S. (2020) Prospective of nanotechnology in degradation of waste water: A new challenges, *Nano-Structures & Nano-Objects*, **22**, 100442.

El-Khateeb, M. & Abdel-Shafy, H. (2022) Selected constructed wetlands case studies in Africa. In: *Asian and Latin American Countries. Constructed Wetlands for Wastewater Treatment in Hot and Arid Climates* (Stefanakis, A., eds). Springer, Cham, pp. 379–394. https://doi.org/10.1007/978-3-031-03600-2_21.

Emenike, C., Tenebe, I., Omole, D., Ngene, B. U., Oniemayin, B. I., Maxwell, O. & Onoka, B. I. (2017) Accessing safe drinking water in sub-Saharan Africa: Issues and challenges in South–West Nigeria, *Sustainable Cities and Society*, **30**, 263–272.

Ettalouia, Z., Salaha, S., Aguelmousb, A., Jadac, A. & Taleb, A. (2021) Use of ferric chloride contained in the rejects from the steel industry as a coagulant for the fuel washing wastewater treatment, *Desalination and Water Treatment*, **234**, 68–76.

Fouda, A., Hassan, S. E.-D., Saied, E. & Azab, M. S. (2021) An eco-friendly approach to textile and tannery wastewater treatment using maghemite nanoparticles (γ-Fe_2O--NPs) fabricated by *Penicillium expansum* strain (K-w), *Journal of Environmental Chemical Engineering*, **9** (1), 104693.

Jagaba, A. H., Abubakar, S., Lawal, I. M., Abdul Latiff, A. A. & Umaru, I. (2018) Wastewater treatment using alum, the combinations of alum-ferric chloride, alum-chitosan, alum-zeolite and alum-*Moringa oleifera* as adsorbent and coagulant, *International Journal of Engineering Management*, **2** (3), 67–75.

Jhansi, S. C. & Mishra, S. K. (2013) Wastewater treatment and reuse: Sustainability options, *Consilience*, **10**, 1–15.

Jiang, J.-Q. (2015) The role of coagulation in water treatment, *Current Opinion in Chemical Engineering*, **8**, 36–44.

Khilji, S. A., Munir, N., Aziz, I., Anwar, B., Hasnain, M., Jakhar, A. M., Sajid, Z. A., Abideen Z., Hussain M. I., El-Habeeb. A. A. & Yang, H-H. (2022) Application of algal nanotechnology for leather wastewater treatment and heavy metal removal efficiency, *Sustainability*, **14** (21), 13940.

Kumar, A. & Pal, D. (2018) Antibiotic resistance and wastewater: Correlation, impact and critical human health challenges, *Journal of Environmental Chemical Engineering*, **6** (1), 52–58.

Leader, T. T. & Wijnen, M. (2018) *Assessment of Groundwater Challenges & Opportunities in Support of Sustainable Development in Sub-Saharan Africa*. Washington, DC, USA: The World Bank.

Li, N., Ma, J., Zhang, Y., Zhang, L. & Jiao, T. (2022) Recent developments in functional nanocomposite photocatalysts for wastewater treatment: A review, *Advanced Sustainable Systems*, **6** (7), 2200106.

Liu, X., Yin, H., Zhao, J., Guo, Z., Liu, Z. & Sang, Y. (2021) Understanding the coagulation mechanism and floc properties induced by Fe(VI) and $FeCl_3$: Population balance modeling, *Water Science and Technology*, **83** (10), 2377–2388.

Mainardis, M., Cecconet, D., Moretti, A., Callegari, A., Goi, D., Freguia, S. & Capodaglio, A. (2022) Wastewater fertigation in agriculture: Issues and opportunities for improved water management and circular economy, *Environmental Pollution*, **296**, 118755.

Masi, F., El Hamouri, B., Abdel Shafy, H., Ghrabi Baban, A. & Regelsberger M, A. (2010) Treatment of segregated black/grey domestic wastewater using constructed wetlands in the Mediterranean basin: The zer0-m experience, *Water Science and Technology*, **61** (1), 97–105.

Maurya, S. P., Singh, P. K., Ohri, A. & Singh, R. (2020) Identification of indicators for sustainable urban water development planning, *Ecological Indicators*, **108**, 105691.

Miarov, O., Tal, A. & Avisar, D. (2020) A critical evaluation of comparative regulatory strategies for monitoring pharmaceuticals in recycled wastewater, *Journal of Environmental Management*, **254**, 109794.

Rajagopal, R., Choudhury, M. R., Anwar, N., Goyette, B. & Rahaman, M. S. (2019) Influence of pre-hydrolysis on sewage treatment in an Up-Flow Anaerobic Sludge BLANKET (UASB) reactor: A review, *Water*, **11** (2), 372.

Regelsberger, M., Baban, A., Bouselmi, L., Abdel-Shafy, H. & El Hamouri, B. (2007) Zer0-M, sustainable concepts towards a zero outflow municipality, *Desalination*, **215**, 64–72.

Wang, H., Wang, T., Zhang, B., Li, F., Toure, B., Omosa, I. B., Chiramba, T., Abdel-Monem, M. & Pradhan, M. (2014) Water and wastewater treatment in Africa – Current practices and challenges, *CLEAN – Soil, Air, Water*, **42** (8), 1029–1035.

Xu, J., Yang, H., Fu, W., Du, K., Sui, Y., Chen, J., Yi, Z., Li, M. & Zou, G. (2007) Preparation and magnetic properties of magnetite nanoparticles by sol–gel method, *Journal of Magnetism and Magnetic Materials*, **309** (2), 307–311.

Xu, J., Yang, L. & Zhou, X. (2023) A systematical review of blackwater treatment and resource recovery: Advance in technologies and applications, *Resources, Conservation & Recycling*, **197**, 107066.

Zehra, A., Upadhyay, R. & Ahmad, A. (2019) Biological treatment of waste: An overview, In: *Advances in Waste-to-Energy Technologies*, Boca Raton: CRC Press, pp. 95–113.

Zhongming, Z., Linong, L., Xiaona, Y., Wangqiang, Z. & Wei, L. (2020) *Sanitation and Wastewater Atlas of Africa*. African Development Bank, United Nations Environment Programme, GRID-Arendal.

Zhou, Y., Guo, B., Zhang, L., Zou, X., Yang, S., Zhang, H. X., Xia, S. Q. & Liu, Y. (2020) Anaerobically digested blackwater treatment by simultaneous denitrification and anammox processes: Feeding loading affects reactor performance and microbial community succession, *Chemosphere*, **241**, 125101.

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