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# Facile synthesis of Ca-(OH)<sub>2</sub>-Mg nanocomposite and its applications for the removal of manganese from drinking water: A pilot study

Kgolofelo Nkele<sup>1,2\*</sup>, Lizzy Mpenyana-Monyatsi<sup>1</sup>, Vhahangwele Masindi<sup>1,2,3</sup>

<sup>1</sup>Department of Environmental, Water and Earth Science, Faculty of Science, Tshwane University of Technology, Private bag X680, Pretoria 0001, South Africa

<sup>2</sup>Magalies Water, Scientific Services, Research and Development Division, Erf 3475, Stoffberg Street, Brits, 0250, South Africa

<sup>3</sup>Department of Environmental Sciences, College of Agriculture and Environmental Sciences, University of South Africa (UNISA), P. O. Box 392, Florida, 1710, South Africa

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## ABSTRACT

A pilot trial was performed in a potable water treatment plant with a capacity of 16 ML/day. The aim was to determine the removal of manganese using a mechanochemically synthesized Mg-(OH)<sub>2</sub>-Ca nanocomposite. The acquired results were underpinned by state-of-the-art analytical instruments. Specifically, the trials were performed for 157 hr using hydrated lime, periclase, and their nanocomposite individually. The key performance indicators were manganese, turbidity, electrical conductivity (EC), and pH. The results showed an increase in pH from  $\pm 7.46$  to  $\geq 7.5$ ,  $\geq 8.2$ , and  $\geq 7.8$  and EC from  $\pm 0.24$  to  $\geq 0.28$ ,  $\geq 0.57$ , and  $\geq 0.58$  mS/cm for hydrated lime, periclase, and their nanocomposite, respectively. Manganese was reduced from  $\pm 400$  to  $\leq 80$   $\mu\text{g/L}$ ,  $\leq 89$   $\mu\text{g/L}$ , and  $\leq 54$   $\mu\text{g/L}$  for hydrated lime, periclase, and their nanocomposite, respectively. The NTU was reduced to  $\leq 1$  for all the chemicals but registered the following sequence:  $\leq 0.40$ ,  $\leq 0.85$ , and  $\leq 0.89$  for hydrated lime  $\geq$  nanocomposite  $\geq$  periclase, respectively, from 6.45 NTU. The findings of this study demonstrated the capabilities of nanomaterials in increasing the pH of the product solution and attenuating manganese and turbidity to the required levels. Lastly, the material costs denoted R 6300.00 (323.98 USD)/week for the nanocomposite, and this was cheaper when compared to individual materials. Interestingly, the nanocomposite denoted superior and cost-effective performance compared to individual materials and will be a great success for the attenuation of manganese and other contaminants, hence enhancing its ferocious versatility in water treatment.

\*Corresponding author Tel.: 0722859461

E-mail: [kgolofelon@magalieswater.co.za](mailto:kgolofelon@magalieswater.co.za)

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## 1. Introduction

Manganese is a metallic element with reddish-grey colour. It is the twelfth most copious element in the earth's crust [1]. An increase in the degradation of aqueous environments by manganese is grossly linked to anthropogenic activities, i.e., industrial, municipal, and agricultural processes [2]. As such, the release of effluents containing elevated levels of manganese to the receiving ecosystem that is not able to sequester such a large amount of manganese naturally has negatively affected the environment and fiercely deteriorated the quality of water, ecological systems, and aquatic health [3]. According to toxicological studies and epidemiological reports, concentrations above the tolerance limit can cause numerous problems [4]; they also grossly compromise disinfection effectiveness, pipe clogging [5], supply stains on fixtures, and result in beverages having a taste that is astringent, undesirable, and bitter or medicinal [6-7]. On consumption, it can result in public health issues [8], such as aggravated methaemoglobin [5]. In light of the enumerated challenges, manganese must be removed from water intended for drinking ahead of exposure to living organisms, specifically in regard to the aforementioned reasons that are of great concern [9]. Stringent regulatory frameworks have set a maximum allowable limit for manganese at 400 µg/L in drinking water [10] to ensure the protection of the environment [11]. The United States Environmental Protection Agency (USEPA) has set 300 µg/L as a health limit and 50 µg/L as an aesthetic limit for manganese in water intended for drinking [12]. As such, the final product water has to comply with set drinking water standards and specifications to avoid exposing consumers to health and aesthetic risks [13]. As a countermeasure, diverse technologies have been developed, piloted, and implemented to remove manganese from drinking water; they include but are not limited to ozonation [14], electrochemical treatment [15], chemical precipitation [16], filtration, i.e., reverse osmosis [17], adsorption [18], and biological processes [19]. These methods can be effectively used for removing manganese from water, but there are issues with these techniques that require attention. In particular, adsorption is not practically viable for bigger

operations like drinking water plants due to regeneration requirements [20]. Filtration and ion exchange generate brine that is difficult to dispose [21]. Oxidation with chlorine has proven to be effective in manganese removal, as manganese (IV), but has limited capacity for concentrated solution [22]. For decades, chemical precipitation has been broadly utilized for manganese removal from drinking water and other wastewater streams. Hydrated lime is mainly used for pH balance and the precipitation of chemicals as hydroxides [23]. Manganese is one of the chemicals precipitated by hydrated lime. Mulyadi et al. [24] evaluated the effect of aluminum sulfate and calcium oxide on the removal of colour, manganese, and iron at Peat WTP. Their results indicated 100% manganese removal. Significant removal of manganese using hydrated lime has also been reported in the literature [25-29]. Magagane et al. [30] evaluated the precipitation of manganese from aqueous solution by utilizing activated or calcined magnesite, i.e., MgO. Similar results have been reported in the literature [31-34]. Furthermore, dolomite, a mineral rich in magnesium and calcium carbonate, has been utilized for the removal of manganese but with limited capacity due to the carbonate fraction buffering pH increasing ability [35-36]. However, its calcined derivative has not been explored for removing manganese from water. Our previous laboratory study investigated manganese removal from drinking water using a mechanochemically activated Ca-Mg-hydrated nanocomposite. The obtained results confirmed ≥83% removal efficacy for manganese at pH ≥11.35. As such, the current study seeks to upscale the laboratory set-up to a full-scale implementation. Considering the above-mentioned, this study was conceptualized to assess the effectiveness and robustness of an Mg-(OH)<sub>2</sub>-Ca nanocomposite on a pilot scale, using 16 ML/day drinking WTP. Additionally, material costs were pursued to demonstrate the viability of the technology, specifically focusing on the chemical costs. This study will go a long way in minimizing the over-exploitation of resources by coming up with cheaper and synergistic chemicals for removing manganese in water intended for drinking, thriving to enhance cleaner production processes.

**2. Materials and methods**

**2.1. Acquisition of chemicals, samples, and reagents**

Hydrated lime and periclase (MgO) were procured from Protea Chemicals (Pty) Ltd. Real River water was used in this pilot study. The experiments were performed in a 16 ML/day drinking water treatment plant, which received raw water from the Wilge River.

**2.2. Fabrication of the nanocomposite**

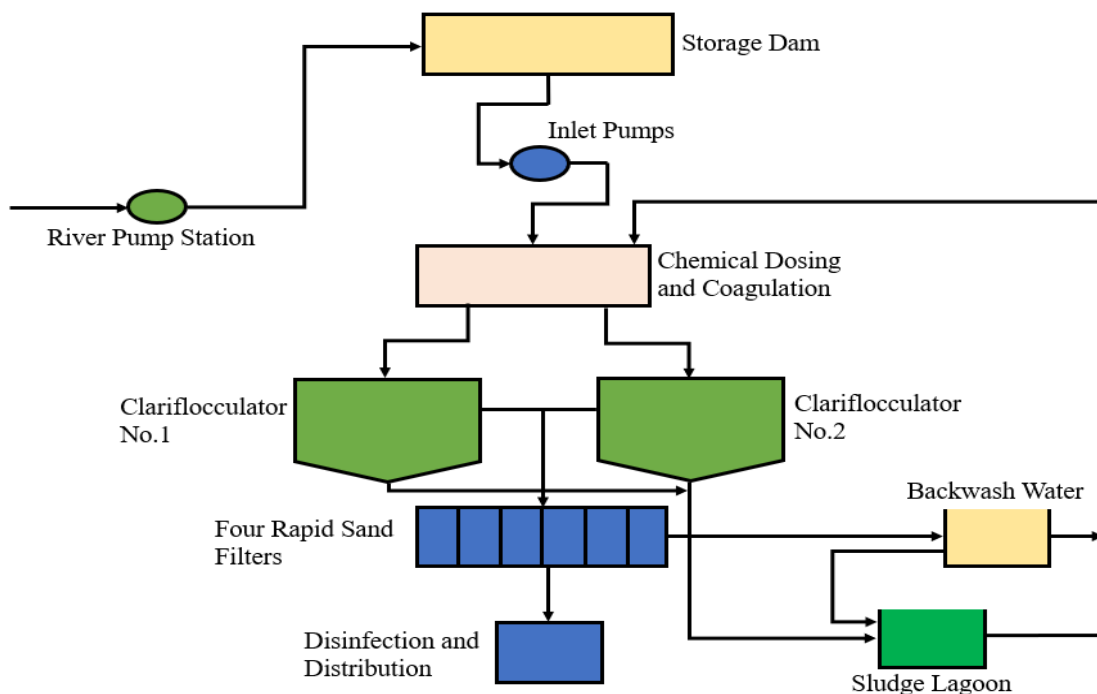
For the purpose of this pilot study, a mechanochemically synthesized nanocomposite was utilised for removing manganese from river water. A similar method proposed by Hu and Zhang [37] was adopted where they mechanochemically prepared a mineral based adsorbent and evaluated its effectiveness in purification ability for wastewater. Calcite was mixed with other materials (composite of calcite with ferrous salt, composite of calcite with zero-valent iron). Khusnutdinov and Isupov [38] performed a hydroxycarbonate form of layered magnesium aluminum hydroxides mechanochemical synthesis. Bester et al. [39] mechanochemically synthesized layered double hydroxides. A mill was used in this procedure [37-39]. In this research, the Mg-(OH)<sub>2</sub>-Ca nanocomposite was synthesized using a

vibratory ball-miller. The MgO-NPs and hydrated lime were mixed at 1:1 (w/w) wt.% mass ratios. The material mixture was then crushed and homogenized for 30 minutes at 1600 rpm using a vibratory ball mill until fine powder particles were acquired. After milling, the homogeneous sample was sieved ≤32 μm size of particles. Subsequently, the samples were packaged and stored in 25 kg bags until needed for removing manganese from drinking water.

**2.3. Water purification process**

In this pilot study, typical water treatment processes were used, including pre-chlorination, coagulation, flocculation, pH correction, clarification, gravity sand filtration, and post-chlorination. The stage-wise water purification process is exemplified in Figure 1.

As shown in Figure 1, manganese rich water was introduced into the water treatment plant; thereafter, the water was transferred to the mixing tank using a centrifugal pump for pre-chlorination by chlorine gas. At the same point, polyelectrolyte was added. After the addition of the polymer for the coagulation process, alkalis, namely MgO, hydrated lime, and Mg-(OH)<sub>2</sub>-Ca nanocomposite, were added to the upwelling chamber for rapid mixing. This was done for individual materials.



**Fig. 1.** A schematic diagram of the followed water purification process.

The water then flowed into a v-shaped Dortmund clarifier with a mixer at the center for floc formation. After the formation of flocs, the water was then clarified, and the suspended particles, including manganese colloids, settled. Clean water overflowed on the edge of the cyclic clarifiers while sludge descended to the clarifier bottom. Ultimately, the supernatant, i.e., the overflow, gravitated to the gravity sand filters for further clarification, and the product water was taken to the onsite reservoir (clear well), where post chlorination took place. The desludging process also took place in the clarifier, and the sludge was hydraulically taken to the lagoons via a desludging system, which was situated at the bottom of the clarifier. For the purpose of this trial, the alkalis were varied on a weekly basis. MgO was continuously introduced into the system for seven days on 24 hr operation. Subsequently, hydrated lime was carried out for seven days following the same procedures. Finally, the efficacy of their nanocomposite was also evaluated in the plant for seven days and 24 hr. The dosage was kept between 100–240 g/min, depending on the properties of the raw water. The key performance indicators were turbidity, manganese, pH, and EC. After the experiments, the findings were compared; the obtained results were used to determine the efficacy of individual materials and their nanocomposite at the pilot scale.

#### 2.4. Aqueous characterization

After collection at defined unit processes (raw and final points), the samples were immediately transported to a laboratory, following the standard procedures for sample handling [40–42]. The samples were characterized in the laboratory and accredited according to the South African National Accreditation System (SANAS). The analysis of the samples was carried out within 24 hr from the time they were collected, with regard to physicochemical properties and microbiological indicators connected with the bacteria of faecal genesis. A brief explanation of the equipment utilized and the analytical methods used to evaluate the samples are specified as follows. AQA (analytical quality control) and QA (quality assurance) evaluations of the research laboratory were carried out utilizing the standards of the US

NIST (National Institute of Standards and Technology) and the techniques that are established in the laboratory, which are accredited by SANAS. Lastly, accredited techniques, procedures, and standards were carried out for the collection of the samples and their analysis (ex-situ and in-situ), as outlined in the APHA (American Public Health Association) guidelines [40–42].

#### 2.5. Characterization of physicochemical properties

A variety of analytical instruments were used to determine the physicochemical parameters of the scrutinized aqueous samples. Chromium, aluminum, colour, manganese, iron, calcium, and magnesium were ascertained using ICP-MS (Inductively coupled plasma mass spectrometry), XSeries 2, ICP-MS, which was provided by Thermo scientific from Hanna-Kunath-Str. 11 28199 Bremen, Germany. The ICP-MS was joined together with an ASX-520 Auto sampler and ICP-OEM (inductively coupled plasma-optical emission spectrometry), 5110 ICP-OES vertical dual view, Agilent Technologies Australia, manufactured in Malaysia. The ICP-OES was combined with the Agilent SPS 4 Autosampler. The nitrate, sulphate, and hardness were ascertained using the Gallery plus photo spectrometer, an automated chemistry analyzer, provided by Thermo Fisher Scientific, manufactured in Finland (Vantaa). The EC, pH, and TDS were ascertained by using a HANNA Multiparameter probe. HI9828 Multiparameter Water Quality Portable Meter. EC and pH were determined at 25 °C. Lastly, the turbidity was measured using a TL23 series laboratory turbidimeter with LED light to ensure high range turbidity. Geosmin was determined using a TQ series gas chromatography-mass spectrometry (GC-MS) manufactured by Dow Chemical, Midland, Michigan.

#### 2.6. Microbiology

The estimation of the concentrations of E. coli and total coliform was done using the guidelines obtained from the APHA guidelines. To be specific, measurements were assessed using the MF (membrane filtration) technique, whereby the sample was passed via a cellulose filter with a pore size of 0.45 µm. Once the filtration process was completed, the samples were cultured on mediums

of agar followed by incubation at 36°C for up to 24 hr. The concentrations of bacteria were then determined [40,42]. Algae was also determined following the APHA guideline, where the MF technique was also used, followed by microscopy for enumeration and identification.

### 2.7. Material costs

The process material costs were determined for all chemicals utilized in the WTP for the purpose of quantifying the cost of the aqueous solution treatment using the developed nanocomposite and individual materials or feedstocks. In this assessment, the maintenance, equipment, labour, and electrical costs were not included to simplify the total costs. The focus of the assessment was only on the usage of chemicals to achieve the removal of manganese from river water. In essence, this study only focused on the OPEX of the system, specifically the pH control reagents and their financial impacts on the operation of the water purification plant.

## 3. Results and discussion

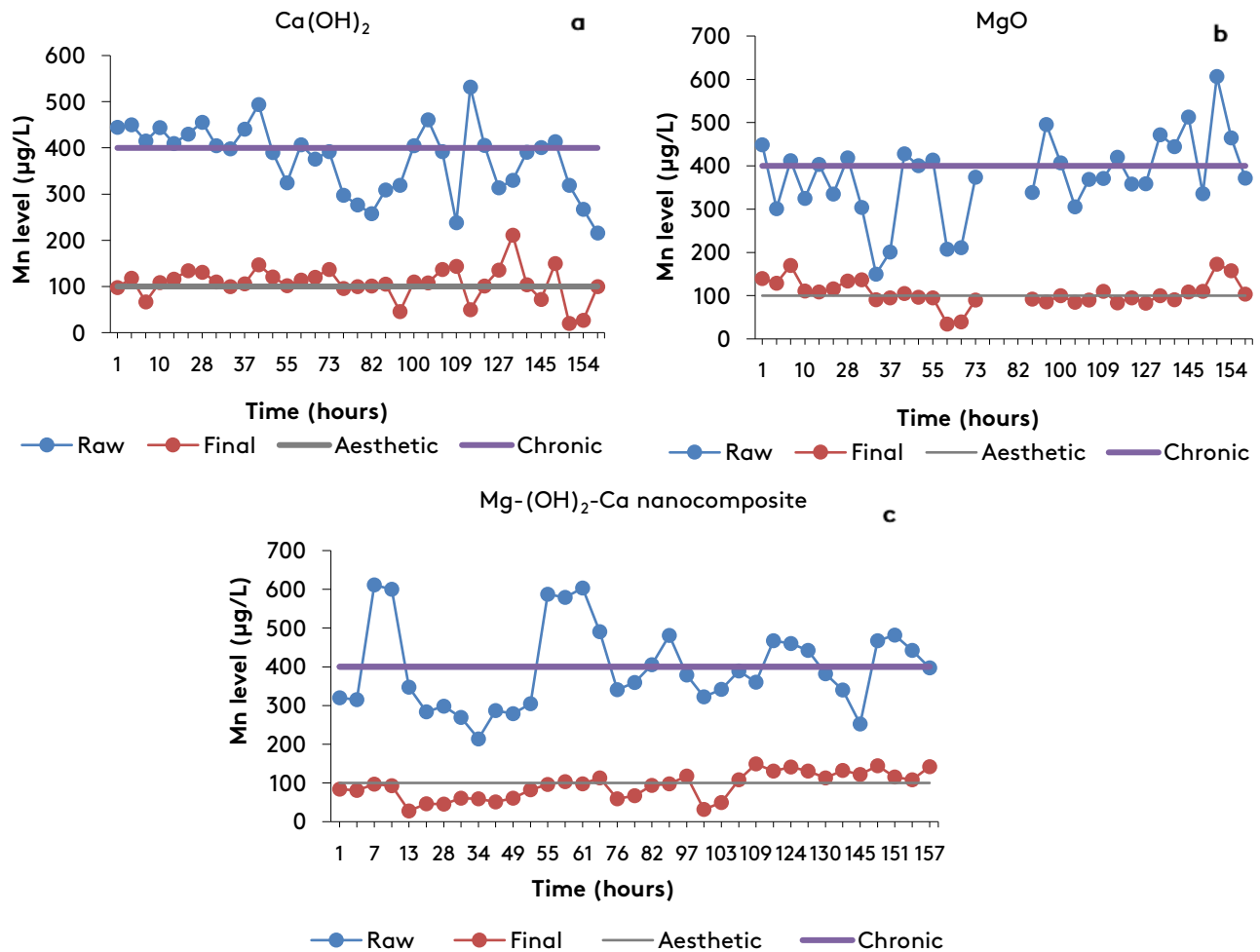
### 3.1. Pilot studies

The effect of hydrated lime ( $\text{Ca}(\text{OH})_2$ ), periclase ( $\text{MgO}$ ), and their nanocomposite on manganese levels, turbidity, EC, and pH are reported in this section. The experiments were performed at a 135 L/s plant flow and 100-240 g/min feedstock dosage. Specifically, the experiments were conducted for 21 days.

### 3.2. Effect of alkaline agents on manganese removal

Variations in manganese levels as a function of hydrated lime ( $\text{Ca}(\text{OH})_2$ ), periclase ( $\text{MgO}$ ), and their nanocomposite are shown in Figure 2. Individual experiments were carried out for seven days, and the results were reported on a three-hour basis.

Figure 2(a-c) represents the trend of manganese during the treatment of water for 157 hr with the hydrated lime,  $\text{MgO}$ , and  $\text{Mg}(\text{OH})_2\text{-Ca}$  nanocomposite. The aesthetic limit for manganese is  $\leq 400 \mu\text{g/L}$ , while the chronic health limit is  $\leq 100 \mu\text{g/L}$  in accordance with SANS 214:2015 [43]. During water treatment, it was observed that manganese decreased from 400 to  $\leq 100 \mu\text{g/L}$  ( $\geq 75\%$  removal efficacy) from raw to final in most of the hours. A similar trend was observed throughout the experiments of the pilot duration. When comparing the individual materials, the nanocomposite confirmed its outstanding performance; the  $\text{MgO}$  performed better than hydrated lime in terms of manganese removal. Interestingly, all the chemicals managed to reclaim water to the required standard for both chronic and aesthetic risk determinants. However, slight fluctuations in the removal efficacy could be noted, but the results were consistent during the duration of the trial. Similar results were obtained by Abdel-Shafy [25], who investigated chemical treatment for heavy metal removal from industrial wastewater. Lime was one of the chemical coagulants used for heavy metals precipitation. The results indicated that by increasing pH with lime, a 93% removal rate of manganese was achieved. Oustadakis et al. [44] investigated the precipitation of cobalt and nickel from sulphate leach liquor by utilizing  $\text{MgO}$  pulp as a neutralization agent. Manganese was one of the impurities identified during the course of their study. The findings depicted an 85% precipitation of manganese at a pH between 8.2-8.8. Navarro and Maiteiz da Matta [45] studied magnesium oxide application to remove metals in mine water treatment. Their outcomes indicated that using  $\text{MgO}$  effectively reduced manganese from 841 to  $0.6 \mu\text{g/L}$ . Mamakoa et al. [46] compared the performance of  $\text{MgCO}_3$  and  $\text{MgO}$  in treating AMD. They found that by treating the water with  $\text{MgO}$ , manganese decreased from 77.49 to  $16.24 \mu\text{g/L}$ .



**Fig 2(a-c).** Variations in manganese levels in  $\mu\text{g/L}$  as a function of hydrated lime ( $\text{Ca}(\text{OH})_2$ ), periclase ( $\text{MgO}$ ), and their nanocomposite (conditions: 135 L/s flow, 100 - 240 g/min dosage of chemicals, average ambient temperature  $24^\circ\text{C}$ ,  $\text{pH} \approx 7.69$  and  $\text{Mn} \geq 406 \mu\text{g/L}$ ).

### 3.3. Effect of alkaline agents on turbidity

Variations in turbidity levels (NTU) as a function of hydrated lime ( $\text{Ca}(\text{OH})_2$ ), periclase ( $\text{MgO}$ ), and their nanocomposite are shown in Figure 3(a-c). Individual experiments were carried out for seven days, and the results were reported on a three-hour basis. Figure 3(a-c) depicts the trend of turbidity during the treatment of water for 157 hr with hydrated lime,  $\text{MgO}$ , and  $\text{Mg}-(\text{OH})_2$ -Ca nanocomposite. The turbidity limit in accordance with SANS 214:2015 is  $\leq 1.00$  NTU [43]. Attaining turbidities that are relatively low in water intended for drinking is a proven indicator for the removal of pathogenic organisms and, therefore, safe drinking water [47]. High turbidity water has been linked to various disease outbreaks, such as gastrointestinal illness [48]. In this regard, it is essential to ensure that water for drinking purposes

consists of turbidities with an acceptable limit. Figure 3(a-c) shows that turbidity decreased significantly for all the chemicals. Specifically, the raw water comprised an average of approximately 6 NTU. However, after treatment, the final turbidity was generally observed to be  $\leq 1.00$  NTU, except for lime in one instance where the NTU was marginally above the threshold. This could be due to higher suspended materials received in the raw water at this hour (127 hr) compared to other hours of treatment. The nanocomposite and  $\text{MgO}$  showed superior performance when compared to hydrated lime. The results obtained from this study corroborated what was stated in a pilot study by Semerjian and Ayoub [49] that alkalized municipal wastewater with lime for treatment. The results indicated  $\geq 99\%$  turbidity removal at  $\text{pH} > 8$ . Another pilot study backing up the results was

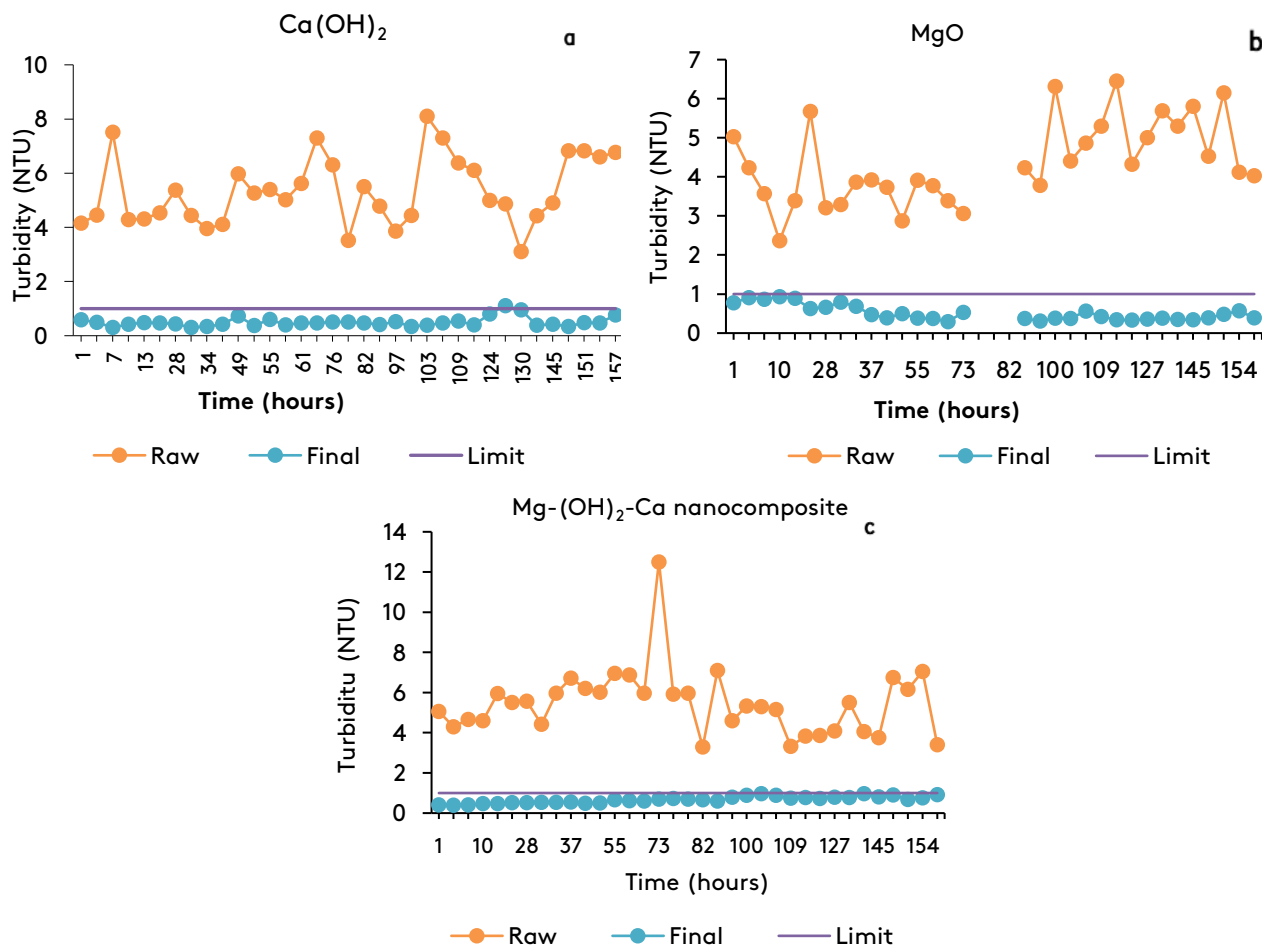


performed by Mirbagheri and Hosseini [50], where lime was used for chemical precipitation. After the water treatment, turbidity was reduced significantly at pH 8.7. Also, a pilot study by Agudosi et al. [51] demonstrated a decrease in turbidity from 35.0 NTU to 4.50 NTU from groundwater when using lime and caustic soda. Prazeres et al. [52] investigated the treatment of wastewater by basic precipitation. A turbidity removal rate of up to  $\geq 98\%$  was observed using hydrated lime. In general, the composite demonstrated the most promising avenue to attenuate the turbidity of the final water product.

### 3.4. Effect of alkaline agents on pH

Variations in pH as a function of hydrated lime ( $\text{Ca}(\text{OH})_2$ ), periclase ( $\text{MgO}$ ), and their nanocomposite are shown in Figure 4(a–c). Individual experiments were carried out for seven days, and the results

were reported on a three-hour basis. Figure 4(a–c) illustrates the trend of pH during the treatment of water with the hydrated lime,  $\text{MgO}$ , and  $\text{Mg}(\text{OH})_2\text{-Ca}$  nanocomposite. The acceptable pH limit, according to SANS 214:2015, is between  $>5$  and  $<9.7$  [43]. As observed in Figure 4(a–c), the pH values increased with the addition of the precipitating agents. Raw water contained an average pH value of 7, which was circumneutral. However, after the addition of alkalis, the pH value of the water increased notably. The final maximum pH value was observed to be  $\geq 8.00$  for all the alkalis. In the pilot setup,  $\text{MgO}$  showed some robust and firm aggressiveness towards pH and other water quality indicators. Be that it may, the pH was not significantly different, even though most literature indicates that effective precipitation of contaminants, including manganese, requires higher pH values ( $\text{pH} \geq 10.0$ ).



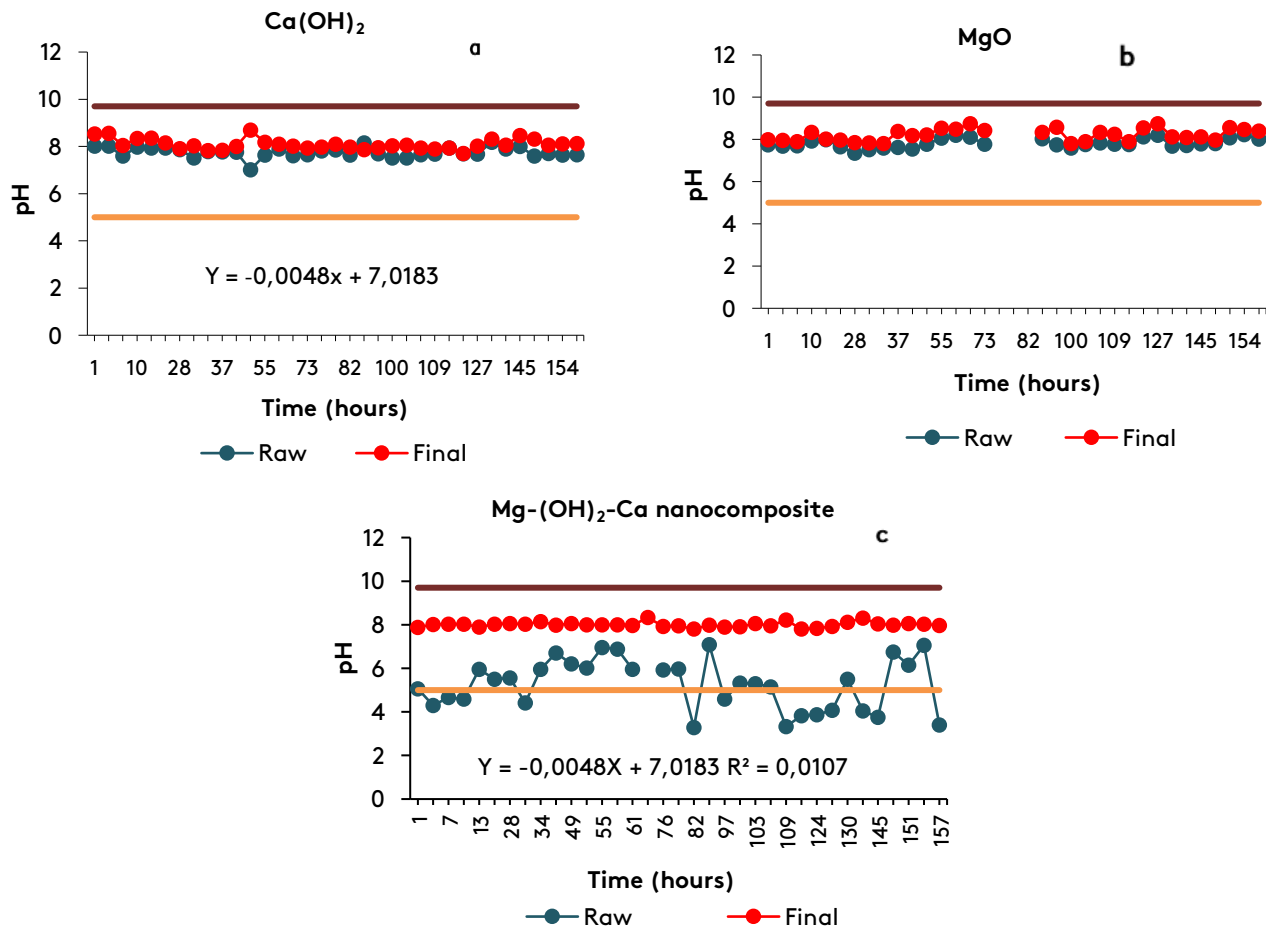
**Fig. 3(a–c).** Variations in turbidity levels (NTU) as a function of hydrated lime ( $\text{Ca}(\text{OH})_2$ ), periclase ( $\text{MgO}$ ), and their nanocomposite (conditions: 135 L/s flow, 100 – 240 g/min dosage of chemicals, average ambient temperature  $24^\circ\text{C}$  and  $\text{pH} \leq 7.69$ , Turbidity  $\geq 5.04$  NTU).

A study by Magagane et al. [30] that utilized magnesite for the treatment of AMD found that a >99% manganese removal rate was achieved at a pH of 10. Aguiar et al. [53] investigated lime along with other materials in removing manganese from AMD and an almost 100% removal rate of manganese was achieved at a pH above 10. Masindi et al. [54] treated mine water by neutralization using different alkali agents. Hydrated lime attained a  $\geq 99\%$  manganese removal at  $\geq 10.9$  pH]; the values obtained by the precipitants used in treating the aqueous solution showed magnificent performance. This finding corroborates what has been conveyed in the following literature. A study by Silva et al. [55] discovered the effective removal of manganese at  $\text{pH} \geq 8.5$  to be 99.9% using limestone. Balladares et al. [56] used lime for coprecipitation and neutralization for treating an acid plant effluent. Their results indicated a manganese removal of  $\geq 99\%$  at a pH between 5

and 9. Masindi et al. [54] evaluated mine water neutralization using different alkali agents. MgO was able to achieve a 100% manganese removal at a  $\geq 9.7$  pH. Last but not least, the nanocomposite always demonstrated an intermediate performance in relation to two individual materials, denoting that it was a derivative of those materials. This chemical could be synergistically put into service for the removal of contaminants from river water.

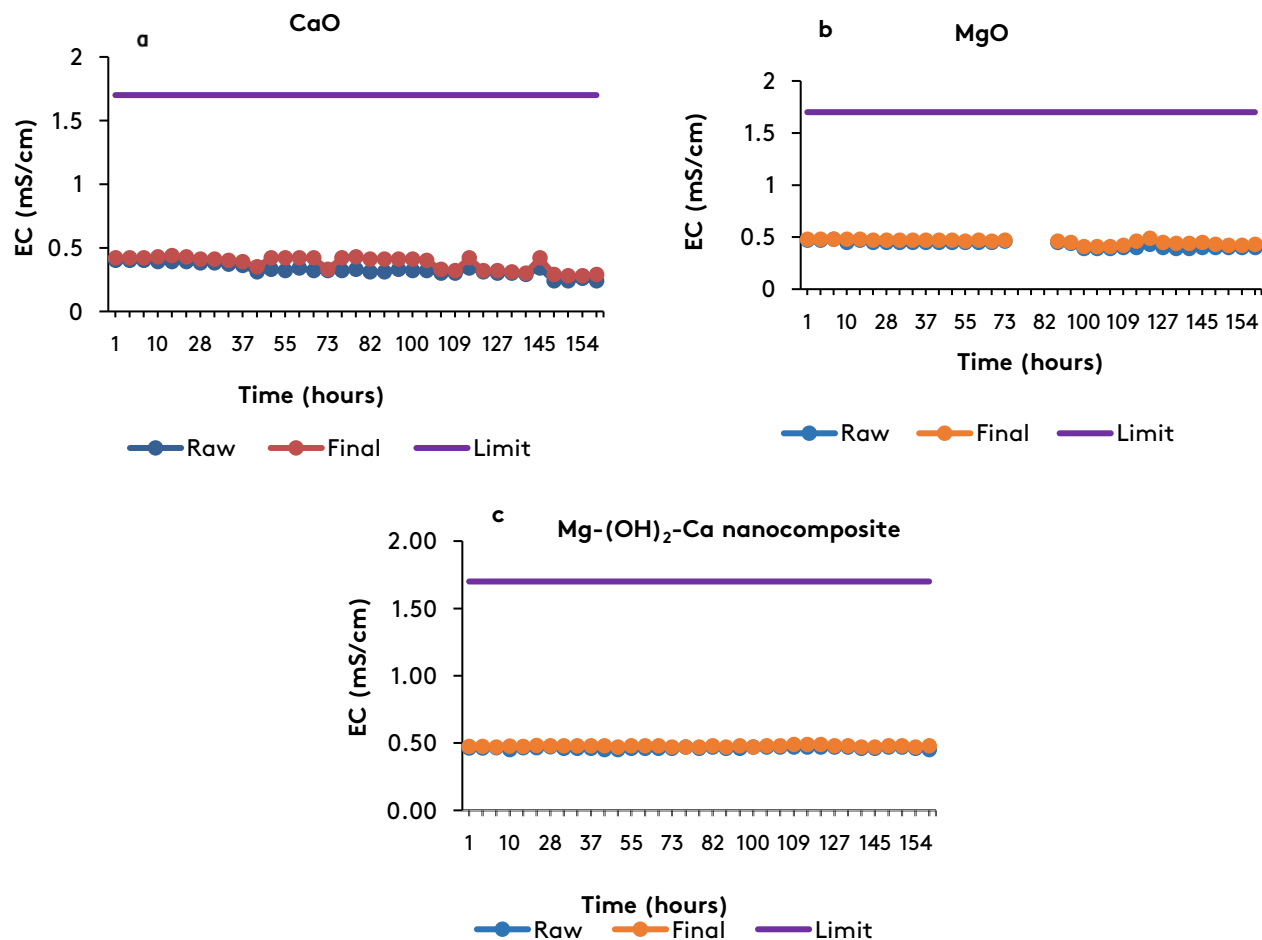
### 3.5. Effect of alkaline agents on EC

Variations in EC as a function of hydrated lime ( $\text{Ca}(\text{OH})_2$ ), periclase ( $\text{MgO}$ ), and their nanocomposite are shown in Figure 5(a – c). Individual experiments were carried out for seven days, and the results were reported on a three-hour basis.



**Fig. 4(a-c).** Variations in pH as a function of hydrated lime ( $\text{Ca}(\text{OH})_2$ ), periclase ( $\text{MgO}$ ), and their nanocomposite (conditions: 135 L/s flow, 100–240 g/min dosage of chemicals, average ambient temperature 24 °C and  $\text{pH} \leq 7.69$ ).





**Fig. 5(a-c).** Variations in EC as a function of hydrated lime ( $\text{Ca}(\text{OH})_2$ ), periclase ( $\text{MgO}$ ), and their nanocomposite (conditions: 135 L/s flow, 100 – 240 g/min dosage of chemicals, average ambient temperature  $24^\circ\text{C}$ ,  $\text{pH} \leq 7.69$  and  $\text{EC} \geq 0.44$  mS/cm).

Figure 5(a-c) shows the trend of EC, which is the capability of the aqueous solution to conduct electricity [57]. EC can be affected by the types of ions, ion concentrations, and the temperature of the aqueous solution [58]. The water was treated for 157 hr with hydrated lime,  $\text{MgO}$ , and  $\text{Mg}-(\text{OH})_2$ -Ca nanocomposite. The EC increased after interacting the reagents with river water, in this case, raw water. Raw water comprised an EC of 0.44 mS/cm on average. The EC for the final water increased for all the precipitating agents except for the lime that dropped. A reduction in EC signified the attenuation of contaminants from the aquasphere, or it suggested a dissolution that was progressive of certain elements from the lime material [45]. Albeit, the increased values complied with the SANS 241-2:2015 regulation that

requires  $\leq 1.70$  mS/cm. Prazeres et al. [52] indicated in their study using hydrated lime for wastewater treatment that high conductivity values were attained at higher pH values (12–13). Navarro and Maetinez da Matta [45] treated mine water using  $\text{MgO}$  and noticed that the contaminated water had an EC of  $<1.0$  mS/cm that increased to  $>7.0$  mS/cm, which was an indication of water mineralization rising sharply.

### 3.6. Treatment of aqueous solution using precipitating agents

The chemical, biological, and microbial properties of raw and product water before and after interacting with  $\text{MgO}$ , hydrated lime, and  $\text{Mg}-(\text{OH})_2$ -Ca nanocomposite is presented in Table 1.

**Table 1.** The chemical, physical, and microbiological properties of feed and product water before and after interaction with hydrated lime ( $\text{Ca}(\text{OH})_2$ ),  $\text{MgO}$ , and  $\text{Mg}-(\text{OH})_2\text{-Ca}$  nanocomposite.

Parameters	Units	SANS 241	Raw aqueous solution	$\text{Ca}(\text{OH})_2$ stage	$\text{MgO}$ stage	$\text{Mg}-(\text{OH})_2\text{-Ca}$ stage
<b>Microbiological determinants</b>						
E.coli	MPN/100mL	$\leq 0$	24	0	0	0
Total coliforms	MPN/100mL	$\leq 10$	3112	0	0	1
Colilert						
<b>Physical determinants</b>						
Colour	mg/L Pt-Co	$\leq 15$	20	5	4	4
EC	mS/cm	$\leq 1.70$	0.24	0.28	0.57	0.58
TDS	mg/L	$\leq 1200$	282	184	0.89	0.85
Turbidity	NTU	$\leq 1.0$	6.45	0.40	0.89	0.85
pH	-	5 to 9,7	7.46	7.53	8.19	7.81
<b>Chemical determinants – macro-determinant</b>						
Nitrate	mg/L N	$\leq 11$	0.28	0.24	0.21	0.21
Sulphate	mg/L $\text{SO}_4$	$\leq 500$	11	10.4	85	82.4
<b>Chemical determinants – micro-determinant</b>						
Iron	$\mu\text{g/L Fe}$	$\leq 300$	476	3.29	0.37	0.37
Mn (manganese)	$\mu\text{g/L Mn}$	$\leq 400$	342	80	89	54
Aluminium	$\mu\text{g/L}$	$\leq 300$	609	27	55	49
Chromium	$\mu\text{g/L Cr}$	$\leq 50$	0.6	0.6	0.06	0.06
<b>Natural chemicals</b>						
Geosmin	ng/L	$\leq 10$	174	1	5	3
Algae	cells/mL	$\leq 1000$	7411	474	354	137
<b>Protection of infrastructure</b>						
Calcium	mg/L Ca	$\leq 300$	15.8	22.2	32.2	39
Magnesium	mg/L Mg	$\leq 100$	13.2	13	25.8	21
Hardness	mg/L $\text{CaCO}_3$	$\leq 300$	140	129	158	179

In Table 1, the parameters present in the raw aqueous solution were determined before and after the interaction of river water with hydrated lime,  $\text{MgO}$ , and  $\text{Mg}-(\text{OH})_2\text{-Ca}$  nanocomposite. An extension of analysis was done to determine the manganese and other parameters removed in water. The parameters determined were compared to the SANS 241-2:2015 drinking water quality standards [43]. Numerous parameters exceeded the water quality standards set for drinking water before treatment, i.e., iron, manganese, turbidity, aluminium, colour, geosmin, algae, E. coli, and total coliform colilert; however, all parameters complied after treatment. In order to obtain the optimum pH value for manganese and other contaminants removal, precipitants were utilized for the aqueous solution treatment. Adding the precipitating agents increased the pH from 7.46 to 7.53, 8.19, and 7.81 by the hydrated lime,  $\text{MgO}$ , and  $\text{Mg}-(\text{OH})_2\text{-Ca}$  nanocomposite, respectively. All reagents increased the pH, which satisfied the precipitation process of manganese and other contaminants. After treatment, manganese was

recorded at 80  $\mu\text{g/L}$ , 89  $\mu\text{g/L}$ , and 54  $\mu\text{g/L}$  for hydrated lime,  $\text{MgO}$ , and  $\text{Mg}-(\text{OH})_2\text{-Ca}$  nanocomposite, respectively. Geldenhuys et al. [29] also confirmed the removal of manganese with the lime process. In this study, the results indicated that manganese was removed from 17 mg/L to 0.01 mg/L at an 8.50 pH value. In another evaluation piloted by Yan et al. [59], lime was used for the precipitation of metals and indicated that the removal rate for manganese was  $\geq 99\%$  at pH 8.0. Iron was drastically reduced from 476  $\mu\text{g/L}$  to 0.37  $\mu\text{g/L}$  by both  $\text{MgO}$  and the  $\text{Mg}-(\text{OH})_2\text{-Ca}$  nanocomposite, whereas hydrated lime reduced it to 3.29  $\mu\text{g/L}$ . This is backed up by a study by Liu et al. [60], who utilized  $\text{MgO}$  for iron removal in wastewater. The outcomes indicated  $\geq 99\%$  iron removal rate. Vlyssides et al. [61] also investigated the effect of lime on iron and sulphate removal in wastewater. The results indicated a  $\geq 82\%$  iron removal efficacy at a pH of 7.0. Yan et al. [59] also depicted a 100% iron removal efficacy at a pH of 4 during AMD treatment using lime. Brbootl et al. [62] used  $\text{MgO}$  and lime for the chemical

precipitation of metals, including iron. The removal rate was obtained at  $\geq 99\%$  with a pH of 5 for iron when using both precipitants. The low pH for iron precipitation was due to the metal's lowest solubility being at a pH between 4 and 5 [62]. The turbidity of the aqueous solution decreased from 6.45 NTU to 0.40 NTU, 0.89 NTU, and 0.85 NTU by the hydrated lime, MgO, and Mg-(OH)<sub>2</sub>-Ca nanocomposite, respectively. They also reduced colour from 20 mg/L Pt-Co to 5 mg/L Pt-Co, 4 mg/L Pt-Co, and 4 mg/L Pt-Co and aluminium from 609  $\mu\text{g/L}$  to 27  $\mu\text{g/L}$ , 55  $\mu\text{g/L}$ , and 49  $\mu\text{g/L}$ . An investigation by Georgiou et al. [63] indicated the effectiveness of lime in removing colour. The pH of the water increased to 9.0. A 70-90% removal rate was achieved. Mortula et al. [64] used different coagulants, including lime, to reduce turbidity and other contaminants. Mehmood et al. [65] utilized lime during industrial effluent treatment and the results obtained showed a  $\geq 91\%$  colour,  $\geq 94\%$  turbidity, and  $\geq 93\%$  TDS removal efficiency at pH 11. Uddin et al. [66] used MgO for chemical precipitation, and their results showed that turbidity was reduced to  $\geq 80\%$  at a 0.25gm/250ml dosage and 10.6 pH. The amount of geosmin was recorded at 174 ng/L before treatment, and it was extremely reduced to 1 ng/L, 5 ng/L, and 3 ng/L by hydrated lime, MgO, and Mg-(OH)<sub>2</sub>-Ca nanocomposite, respectively. The microbial parameters were also measured, i.e., algae, E. coli, and total coliform colilert. After treatment, the algae were reduced from 7411 cells/mL to 474 cells/mL by hydrated lime, whereas MgO reduced it to 354 cells/mL and the Mg-(OH)<sub>2</sub>-Ca nanocomposite reduced it to 137 cells/mL. E. coli was not detected after treatment by all the

precipitants, and total coliform colilert was only detected at a value of 1 MPN/100 mL after treatment with the Mg-(OH)<sub>2</sub>-Ca nanocomposite. The following literature supports these outcomes. A pilot study by Ewerts et al. [67] investigated a WTP that used hydrated lime as one of their chemicals; they found that the coagulation process in which lime was added could enhance the effectiveness of algae removal. Another study by Polaczyk et al. [68] utilized hydrated lime to stabilize freshwater sediments by reducing the microbiological population. The outcomes indicated that the total coliforms and E. coli decreased to  $\geq 99\%$  at pH 12. EC showed an increase after treatment by all precipitating agents. Even though the EC increased, it never passed the drinking water limits when compared to that of SANS 241-2:2015 for all agents. The same applied to calcium, hardness, magnesium, nitrate, sulphate, chromium, and TDS. When comparing the chemical precipitating agents, the Mg-(OH)<sub>2</sub>-Ca nanocomposite showed greater performance compared to individual materials with regard to manganese and other contaminants removal due to its properties obtained from hydrated lime and MgO.

### 3.7. Material costs

The process of removing manganese from river water was examined from a cost viewpoint. The estimation of the material costs was based on the chemical reagent type in accordance with the chemical dosages in the pilot study. In essence, this evaluation focused solely on the utilized chemicals. Costs, which included maintenance, labour, electricity, design, etc., were excluded. The results for the material costs are depicted in Table 2.

**Table 2.** Material costs (21 days) for hydrated lime, periclase, and their nanocomposite.

Items	Unit cost	Quantity of item	Units	Total
Hydrated lime, R/ton	R2500	2800	Kg	R7000 (359.98 USD)
Magnesium oxide, R/ton	R3500	3150	Kg	R11025 (566.97 USD)
Mg-(OH) <sub>2</sub> -Ca nanocomposite, R/ton	R3000	2100	Kg	R6300 (323.98 USD)
<b>Total cost ®</b>				<b>R24325 (1250.93 USD)</b>

A great number of water treatment processes have been employed for the removal of manganese from aqueous solution. During the pilot scale study, a 16 ML/day water treatment plant was utilized. Table 2 emphasizes the importance of comparing the costs of each ingredient used to determine the most

cost-effective material. The number of bags dosed during the trial was 84 bags of Mg-(OH)<sub>2</sub>-Ca nanocomposite, 112 bags of hydrated lime, and 126 bags of MgO. As indicated in Table 2, the chemicals utilized for the treatment of the aqueous solution contained a commercial value that ranged

between R2500 and R3500. The total cost of all chemicals was R24325 (1250.93 USD). The cost of the required material varied, with hydrated lime, MgO, and the nanocomposite costing R7000, R11025, and R6300, respectively. This was based on the amount of kilograms required to treat the aqueous solution. Using the Mg-(OH)<sub>2</sub>-Ca nanocomposite will reduce the cost compared to the other materials. Therefore, the Mg-(OH)<sub>2</sub>-Ca nanocomposite has sufficient properties compared to the other materials.

#### 4. Conclusions

This pilot-treatment study confirmed the removal of manganese and other contaminants from river water using hydrated lime, MgO, and their nanocomposite (Mg-(OH)<sub>2</sub>-Ca nanocomposite). A plant with a capacity of 16 ML/d was used to fulfil the goals of this investigation. The study employed on-site optimum conditions, which were a 100 mL/min polymer dosage, 1.25 kg/h Pre-Cl<sub>2</sub>, 0.75 kg/h Post-Cl<sub>2</sub>, and 100–240 g/min precipitating agents' dosage. The treatment process successfully offered a strong, high rate, and proven system that could effectively remove contaminants present in wastewater. By using the precipitating agents (hydrated lime, MgO, and Mg-(OH)<sub>2</sub>-Ca nanocomposite), the treatment process promptly attained results of acceptable standards and continually produced water of high quality. The nanocomposite outshined the other individual materials in removing manganese from water as it contained properties of both the hydrated lime and MgO. The results obtained showed that the pH increased from 7.46 to 7.53, 8.19, and 7.81 by the hydrated lime, MgO, and Mg-(OH)<sub>2</sub>-Ca nanocomposite, respectively. Most literature indicates that a pH above 10.0 is required for effective removal of manganese, but the pH values obtained in this study also successfully achieved metal precipitation. Hydrated lime, MgO and Mg-(OH)<sub>2</sub>-Ca nanocomposite reported the removal efficiency of 77%, 74%, and 84% for manganese, respectively; the EC increased in all the precipitating agents after analysis of the samples in an accredited laboratory, but in the trend results, it was decreased by hydrated lime, which could be due to the types of ions, ions concentrations, and the temperature of the

aqueous solution. The trends of the parameters, i.e., manganese, turbidity, EC, and pH, fluctuated throughout the water treatment but depicted a decrease in the contaminants from the raw water through to the final water treatment. The fluctuations could be due to changes in the raw water characteristics (effluent quality), weather conditions (temperature), and plant interruptions. The experiments that were performed indicated that increasing the pH of the water through the use of precipitating agents provided an efficient solution to aesthetic and health problems experienced by consumers. This is supported by our laboratory study, which stated that by increasing the pH of the water using the Mg-(OH)<sub>2</sub>-Ca nanocomposite could achieve a high manganese removal rate [69]. The outcome from the material costs study had a total amount of R24325 (1250.93 USD), with Mg-(OH)<sub>2</sub>-Ca nanocomposite costing less to treat water than the individual materials. Precipitation was a cheaper technique to remove contaminants from wastewater and using it together with the nanocomposite as an alkaline agent could be very cost-effective. This investigation can be of great assistance to many WTPs with regard to manganese removal from river water. However, it is recommended that more studies be undertaken in the future regarding the removal of metals in water using the Mg-(OH)<sub>2</sub>-Ca nanocomposite.

#### Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that would influence this paper.

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