Ain Shams Engineering Journal 12 (2021) 4201–4209

Contents lists available at ScienceDirect

Ain Shams Engineering Journal

journal homepage: www.sciencedirect.com

Engineering Physics and Mathematics

Cement kiln dust as an alternative technique for wastewater treatment

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ARTICLE INFO

Article history:
Received 10 January 2021
Revised 12 April 2021
Accepted 24 April 2021
Available online 10 May 2021

Keywords:
Cement kiln dust
Alum
Coagulant
Wastewater treatment
Heavy metals
Environmental alternatives

ABSTRACT

This study was conducted to examine cement kiln dust (CKD) efficiency for wastewater treatment. We analyzed the physicochemical characteristics of wastewater before and after treatment and then we determined its removal potential. The optimum factors of the treatment process were determined using a jar test technique. It was pH (8.1), dosage (1.9 g) and grain size (0.1 mm) with contact time of 30 min at 150 rpm. CKD pollutant removal efficiency reached 85.3, 81.6, 97.1, 86.8, 36, 74, 61.2, and 94.6% for BOD, COD, TP, TN, TDS, salinity, conductivity and turbidity, respectively, with an increase in the concentration of DO of 84%. On the other hand, removal percent of heavy metals achieved were 88.4, 90.9, 88.5, 97.2, 94.2, 70, and 79.9% for Pb, Cd, Zn, Fe, Co, Ni, and Cu, respectively. These results were compared with alum removal potential of wastewater treatment for confirmation. Alum pollutant removal efficiency reached 86.6, 79.6, 96.6, 59.9, 39.7, 65, 59 and 95.2% for BOD, COD, TP, TN, TDS, salinity, conductivity and turbidity, respectively, with an increase in the concentration of DO of 85.3%. On the other hand, removal percent of heavy metals achieved were 82.1, 90.6, 89.1, 96.8, 93.2, 72.8, and 84.1% for Pb, Cd, Zn, Fe, Co, Ni, and Cu, respectively. The methodology carried out in this study indicated that CKD can be used as a good environmental alternative coagulant for low to moderate wasted water as it achieved removal percent similar to that achieved by the common coagulant alum.

1. Introduction

A rapid increase in urbanization and industrial growth has recently been attributed to an increase in the production of different kinds of contaminants such as air pollutants, solid waste and production of wastewater. Industry was the largest contributor to the production of these pollutants, especially heavy industry factories such as cement industry factories [1]. The first cement factory was built in 1911, and then Egypt constructed its first cement company in 1927, so the cement industry is considered an ancient industry in Egypt and the world. At present time in Egypt, there are nineteen cement companies working with forty-two lines of production which are spread throughout Egypt. Although Egyptian cement makers are concerned about the environment, and the cement industry has previously received a lot of research interest to make it less dangerous to the environment [2,3], there are still various wastes produced from its production that require more attention to get rid of it or try to reuse it in a safe way. The cement factories dispose of more than eleven million tons/year solid waste and produce more than 1.3 million tons/year wastewater [4]. These materials are not recycled or reused in industry as raw materials or synthetic fuels and most of them are disposed of in a landfill. Uncontrollable disposal of these pollutants increases environmental pollution problems [5,6]. The most important material disposed of in these materials is cement kiln dust (CKD).

Nowadays, recycling of industrial waste products has been successively experienced in mitigation of the environmental challenges in sustainable and economic strategies by solving an environmental problem and addressing disposal problems. Cement kiln dust (CKD) is considered an important one of these products, especially since it is classified as a non-hazardous by-product of solid waste [7]. It is a heterogeneous by-product dust resulted from cement manufacturing processes with composition resources of carbonate and other raw materials [8]. CKD was introduced back into the clinker-making cycle as a raw material with modern manufacturing techniques. However, the restrictions on the alkali and chloride contents in the cement make it a difficult process. In a pre-

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Peer review under responsibility of Ain Shams University.

Production and hosting by Elsevier

https://doi.org/10.1016/j.asej.2021.04.026

2090-4479/

Nomenclature

CKD	Cement kiln dust	TN	Total nitrogen
BOD	Biological oxygen demand	TDS	Total dissolved solid
COD	Chemical oxygen demand	EC	Electrical conductivity
DO	Dissolved oxygen	P_i	Initial pollutant concentration mg/l
TP	Total phosphorus	P_f	Final pollutant concentration

vious study in the UK, it was reported that over 200,000 tons a year of landfill space could be saved if CKD could be recycled or if alternative uses could be found [9].

According to the previous studies [10,11,12,13,14]. CKD consists of quartz, a small quantity of gypsum and sodium chloride besides limestone, which is the major component of it. It has multiple coagulants (CaO, MgO, Al_2O_3 and Fe_2O_3, respectively), in addition to SiO_2 which makes it a good adsorbent at the same time [15]. Wherefore, CKD has recently been used in wastewater treatment processes. The most popular application of it is for sewage sludge applications, as it is considered a chemical conditioner and stabilizer [16]. It is cheaper than lime and other coagulants and adsorbents so it could reduce the cost of waste treatment with the same performance [17].

Alum (Aluminum sulfate) is probably the most widely used coagulant in water treatment [18]. However, nowadays, aluminium-based coagulant and its use in water treatment has become under examination and inspection. It was reported that alum was found in the water after the treatment at a high level and a large amount of sludge was produced and that raised concern about public health. According to previous studies, neurodegenerative diseases develop if these salts are taken in significant amounts [19].

Regrettably, there is a manifest gap in the previous literature about the usage of cement kiln dust for wastewater treatment and improving its quality. It was concentrated only on the application of it for the removal of heavy metals depending on precipitation/dissolution and adsorption/desorption mechanisms. It was assumed that the predominant mechanism in the removal process is precipitation. Accordingly, this study was confirmed to address this research gap. We examined and evaluated the ability of cement kiln dust usage in water treatment and then compared its performance with one of the most considered and effective coagulants. Alum was selected in this study for the treatment of wastewater and its performance was compared with the environmental alternatives performance under the study (CKD).

This paper first discusses the experimental analysis for the detection of the optimum conditions for the treatment process (batch adsorption mode), using Jar Test, then goes on to analyze the obtained data from the treatment process (mathematic and statistical analysis) and then Explains and interprets results in the discussion section and finally ends with the conclusion section where cement kiln dust confirmed its ability to be used as an alternative technique for wastewater treatment and achieved great potential similar to that of alum in an easy and low-cost way

2. Method and material

Cement Kiln Dust (CKD), was collected from Suez Cement Company, located at K30 Maadi/Ain Sokhna Road, Cairo, Egypt. It was dried for 24 h at 105 °C. The wastewater used in this study was a composite sample of raw water that was collected from different sites along the Nile River-Damietta Branch, (Fig. 1). Sites were chosen according to the study of Hasaballah and Hegazy [20] as the most polluted sites along the branch were chosen. These sites were Eladlia, Bosat Karim Eldein, Talkha and Smnood. The main physicochemical characteristics of the collected water samples were analyzed: Dissolved oxygen (DO), biochemical oxygen demand (BOD), Chemical oxygen demand (COD), total nitrogen (TN), total phosphorus (TP), pH, total dissolved solids (TDS), Salinity, Electrical conductivity (EC) and some heavy metals, (Table 1). Chemical addition, refrigeration, pH control and freezing, were used as preservation methods [21]. The pH value of the samples was measured using (HI 111, HANNA, USA) pH Meter according to Baird [21]. While the turbidity of samples was measured using (Extech TB 400) Portable Turbidity Meter according to Baird [21]. Where Total dissolved solids (TDS), Salinity and Electrical conductivity (EC) were measured by TDS meter (HI 98192 Digital Portable Conductivity/Salinity/TDS meter) as TDS expressed in mg/l and Salinity expressed in dS/m. Dissolved oxygen (DO) in the water sample was detected according to Hasaballah and Hegazy [20], while Chemical Oxygen Demand (COD) and Biological Oxygen Demand (BOD) of the samples were determined according to Hasaballah and Hegazy [22] and Adams [23]. On the other hand, Total Nitrogen was determined according to Rump [24] and the Total Phosphate (TP) value of the samples was detected using Ascorbic Acid Method [21]. Furthermore, the analysis of heavy metals in water samples (Fe, Zn, Pb, Cu, Ni, Cd, and Co) was carried out according to Hasaballah and Hegazy [20]. Fig. 2 describes the research methodology.

2.1. Jar test

It was performed using Stuart Flocculation Jar Tester, 6-place; 230 VAC, 50 Hz (Cole-Parmer, USA). First, we sieved CKD using a Ro tap shaker, **(SS-30 8in Ro-Tap Sieve Shaker)**. Particle sizes of less than 0.1, 0.1, 0.5, 0.7, 1 and 2 mm were selected depending on previous studies, [17,18,19]. The particle size which caused the lowest turbidity was chosen for the follower step. Then we determined the optimum dosage for each coagulant according to Baird [16] and Jagaba and Kutty [25]. Various dosages were added to beakers with paddles. Then the tester turned on a rapid mixing (250 rpm for one minute) then adjusted to the slow mixing (150 rpm for 30 min). After mixing, the flocs were allowed to settle for 30 min. Finally, residual turbidity was measured against dosage using 10 ml pipetted from the highest 5 cm from the surface of each beaker. The optimum dosage gives low turbidity for coagulant. The previous steps were repeated with changes in pH, contact time and grain size for each time to detect optimum pH, contact time and grain size values.

2.2. Wastewater treatment experiments

The optimum dose, pH, grain size and contact time were determined previously, were used for wastewater treatment using coagulation-adsorption technique [26].

by a rapid mix at 150 rpm for one minute followed by a flocculation at 40 rpm for 20 min and a settlement for 30 min [27]. The physical and chemical parameters have been carried out before

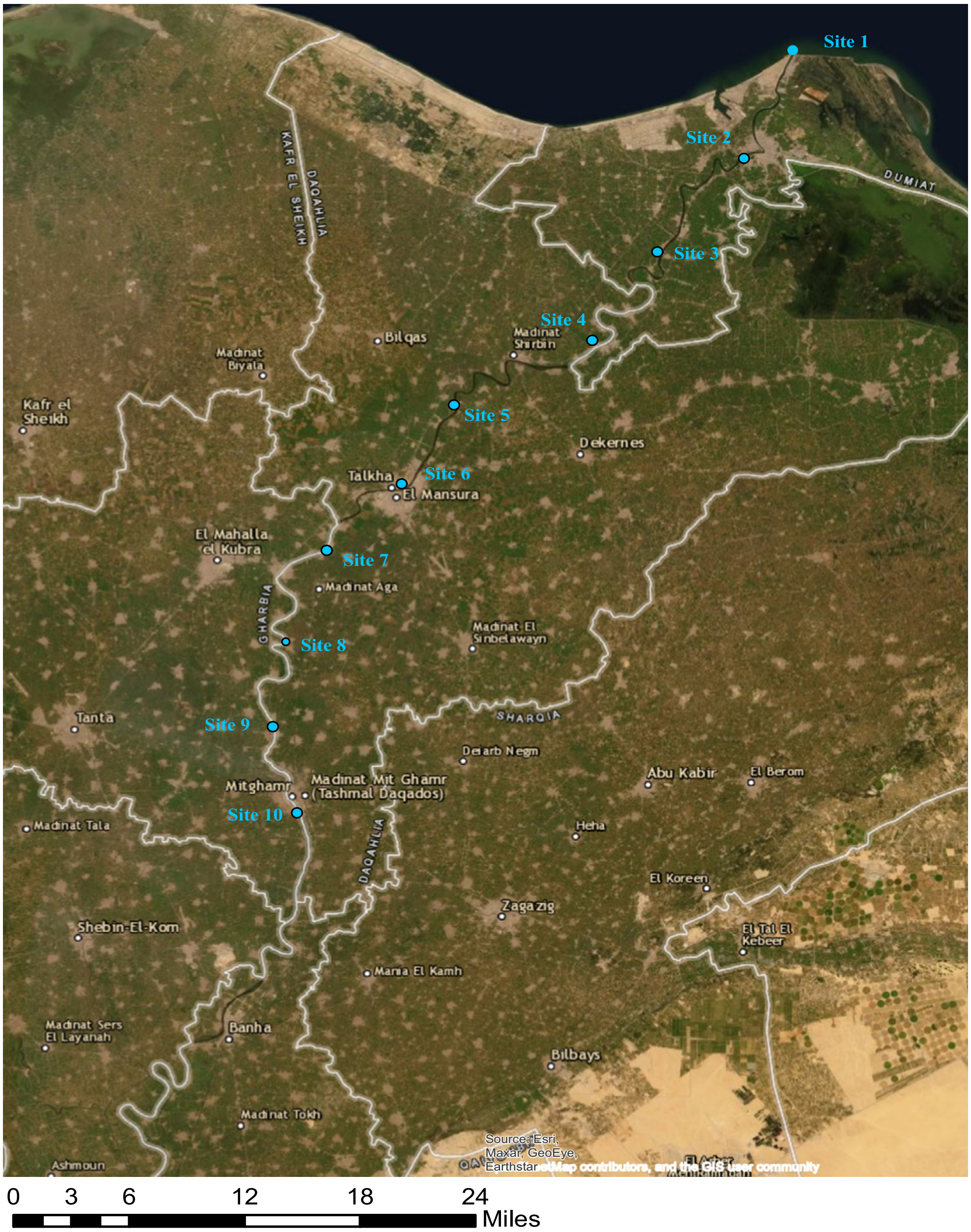

Fig. 1. Geographic Map of the Nile Delta showing the study area (Damietta Branch) and the different ecological sites (1: Ellesan / Ras Elbr; 2: Damietta Dam Region; 3: Eladlia; 4: Shrbas / Faraskoor; 5: Elsero/Elzarqa; 6: Bosat Kareem Eldein / Sherbein; 7: Talkha; 8: Smnood; 9: Meit Ghmr; 10: Kafr Shokr).

and after treatment according to standard methods mentioned before. Concentrations of heavy metals were measured using Inductively coupled plasma mass spectrometry (ICP-MS), **(Nex ION 300 ICP-MS- Perkin Elmer)**.

2.3. Removal efficiency of pollutants

The obtained data was examined to determine the performance of the system. The efficiency of treatment was determined according to the removal percentage of each parameter using the following formula:

$$\text{Removal efficiency } (\%) = (P_i - - P_f)/Pi \times 100$$

where P_i and P_f are the initial and final pollutant concentration [28].

2.4. Statistical analysis

The comparison of means for the two coagulants and the standard deviations were checked for significance ($p \leq 0.05$) using the ANOVA test and the post hoc tests. In addition, the relationships among water parameters were assessed using regression and the correlation coefficient for Pearson. All of these statistical analyses were conducted using SPSS Computer Software, IBM 25.0 Version (SPSS Inc. Chicago, USA).

3. Results and discussion

Sorption and the desorption processes should have optimized operating parameters with a view to achieving the maximum efficient removal with limited resources [29]. The adsorption – coagulation experimental parameters (dosage, pH value, temperature,

Table 1
Treatment with CKD and Alum.

Parameters	Pretreated water mg/l	Post treated with *CKD*		Post treated with *Alum*	
		Conc.	Removal %	Conc.	Removal %
pH	6.9	7.9	–	7.8	–
TS	56.2	4.77	91.5	16.01	71.5
TDS	40.3	25.79	36	24.3	39.7
Salinity	32.7	8.50	74	11.44	65
Conductivity	51.3	19.90	61.2	21.03	59
Turbidity	40.51	2.18	94.6	1.94	95.2
DO	6.53	40.81	84 -	44.42	85.3 -
BOD	23.37	3.43	85.3	3.13	86.6
COD	62.55	11.50	81.6	12.76	79.6
TP	2.8	0.081	97.1	0.086	96.6
TN	1.4	0.184	86.8	0.561	59.9
Cd	0.89	0.089	90.9	0.083	90.6
Cu	4.19	0.842	79.9	0.666	84.1
Pb	3.93	0.455	88.4	0.703	82.1
Fe	19.62	0.549	97.2	0.627	96.8
Co	3.43	0.198	94.2	0.233	93.2
Zn	6.18	0.717	88.5	0.673	89.1
Ni	1.4	0.42	70	0.381	72.8

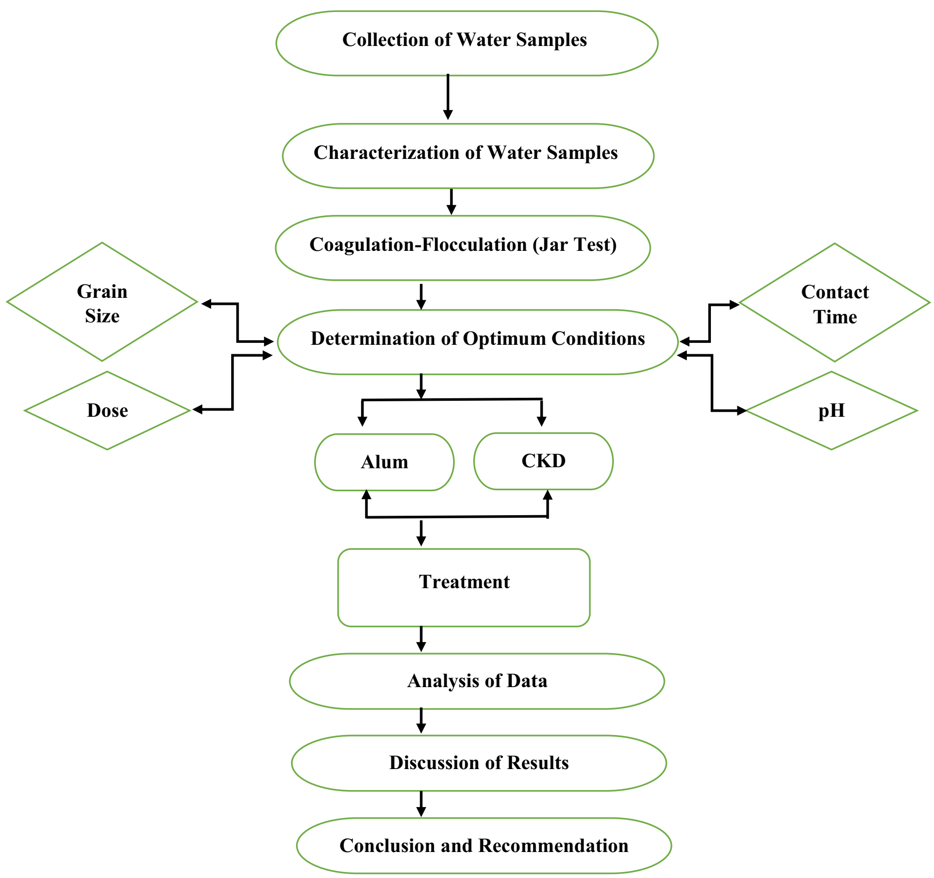

Fig. 2. The research methodology outlines.

retention time, and texture) were optimized for obtaining the optimum treatment condition by using Jar Test.

3.1. Optimum grain size

Fig. 3 represents different grain size removal efficiency as a function of turbidity. The optimum grain size recorded was 0.1 mm for cement kiln dust and was 0.5 mm for alum. These grain sizes achieved the maximum removal of turbidity.

3.2. Optimum dose

The optimum dose of cement kiln dust and alum were determined to be used in treatment, (Fig. 3).

Studying the dosage effect of any adsorbent is economically important as it indicates the efficiency of the adsorbent and the potential of ions to be adsorbed using the lowest dose [30]. The optimum dose was recorded as a function of turbidity through retention time at 150 rpm for 30 min and a temperature of 20°C. It was found that the best removal and improvement of turbidity

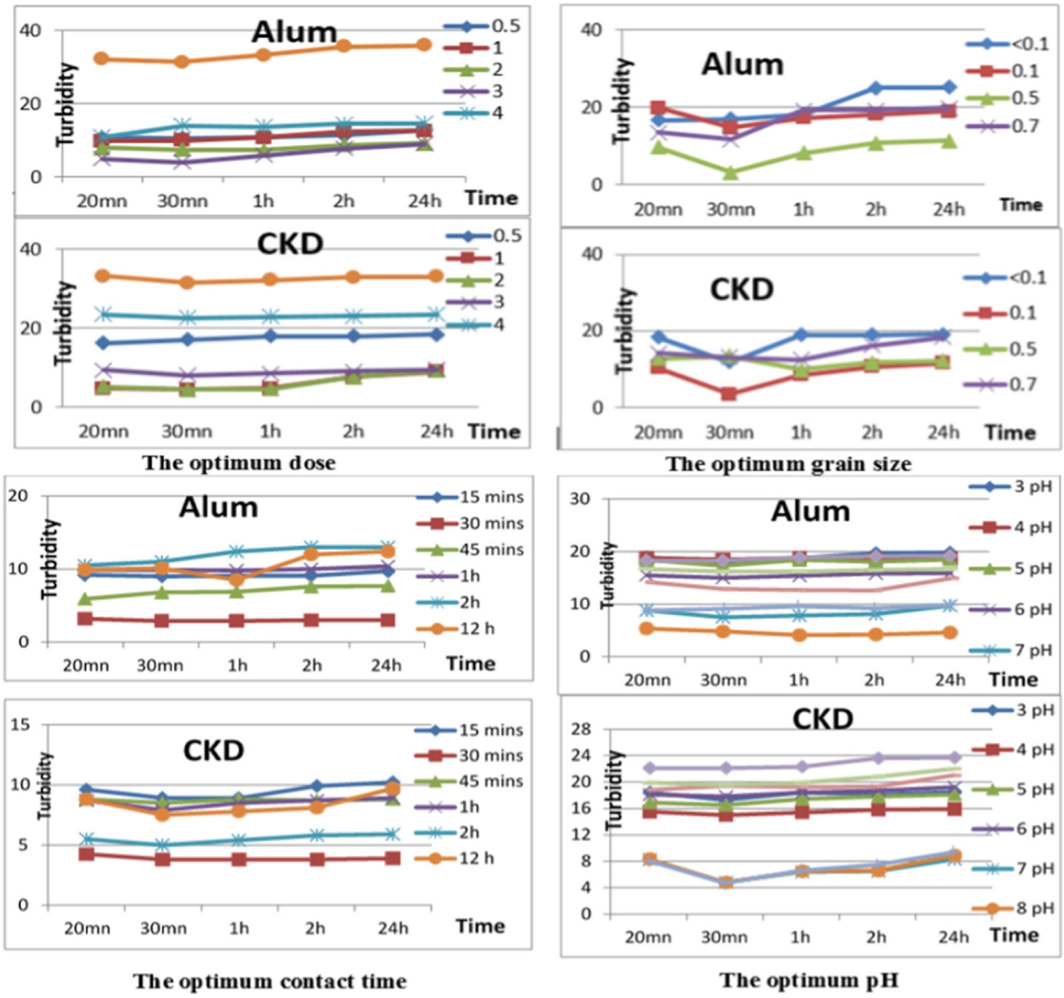

Fig. 3. The optimum conditions (pH, dose, grain size and contact time) for CKD and Alum treatment process.

was achieved at 1.9 g/l for cement kiln dust but it was 3 g/l for alum, (Fig. 3). Particles which result in flocculation are destabilized when coagulants are hydrolyzed. The cationic species and products that are produced from the hydrolysis process are absorbed by particles with negative charges and then they are neutralized. However, it was reported that increasing the dosage at some points has adverse effects and decreases removal efficiency [31]. Jagaba and Kutty [25] investigated alum removal efficiency in water treatment and reported that increasing alum dose increased removal efficiency for most elements while having a negative effect on some metal removal such as Fe metal in their study.

3.3. Optimum pH

After detecting CKD and alum optimum dose, it was used to determine the optimum pH for wastewater treatment. This specific optimum dose was mixed with water samples at a different pH value. The lowest turbidity occurred for alum and cement kiln dust at 8 and 8.1 pH value, respectively, as shown in Fig. 3. This is the result when the pH is set to its natural level. However, at pH values of 6 and 7, the results were quite similar, and increasing the pH value above 9 was not desired.

3.4. Contact time

The contact time was also an important factor in this experiment because it can affect the adsorption kinetics of the adsorbent at a given concentration [32]. Equilibrium was acquired after 30 min, for concentrations of 3 g/l for alum and 2 g/l for cement kiln dust (Fig. 3). The adsorption and removal increased during the first mixing stage and then continually increased with contact time at a slow second mixing stage.

Ghoochian and Panahi [33] thought this could be due to the vacant surface sites which are present in a large number and it is available for contaminant adsorption in the first stage of adsorption. El-Awady and Abo-El-Enein [34] reported that the removal of Mn, Cu, Pb, Cd, Zn, and Cr were 100% at 20 g CKD/l and 150 rpm for 30 min, while the removal of iron and nickel were 98% and 80%, respectively.

3.5. Treatment experiment

The optimum factors defined and used in batch adsorption experiments for the treatment of wastewater were pH (8.1,8), dosage (1.9,3g/l) and grain size (0.1, 0.5 mm) with contact time of 30 min for alum and CKD respectively.

After the treatment, pH was observed to have a slight increase and become in the neutral range with mean values of 7.9 with CKD and 7.8 with alum from the original value of 6.9. This slight increase was supported by Drouiche and Moussaceb [35]. TS removal percent was 91.5% for CKD but it was 71.5% for alum. In the same manner, salinity and conductivity removal percent were 74% and 61.2% for CKD, respectively. While it was 65% and 59%, respectively for alum, (Table 1).

On the other hand, the TDS removal percent was 36% in the case of CKD while it was 39.7% for alum and also for turbidity, the removal percent was 94.6 for CKD and 95.2% for alum and these results were close to results reported by Ahmad and Wong [36], as he stated that, the higher removal efficiency utilizing coagulants (alum) could be assigned by charge neutralization of negatively charged colloidal particles adsorb of positively charged species coagulant, and trap of the colloids in precipitating Al $(OH)_3$ solids.

DO had a high improvement percentage of 84%, which was close to the alum percentage (85. 3%). BOD, COD, TP and TN removal percentages were 85.3, 81.6, 97.1 and 86.8 for CKD, while it was 86.6, 79.6, 96.6 and 59.9, (Fig. 4). The past investigation directed by Ahmad and Wong [36], demonstrated that the COD removal efficiency by Alum doses of 1 g/L was extended from 91% to 91.3%. Mazari and Abdessemed [37] also reported similar results, as he recorded 90% of COD removal percent by using alum. While Kang and Chai [38] reported that, TN removal percent was 38.33% and TP removal percent was 74.29%. Alum is extremely outstanding as an enhancer of phosphorus removal in aerobic biological systems.

Removal percentages of metals like Pb, Cd, Ni, Co, Zn, Cu and Fe were 88.4, 90.9, 70, 94.2, 88.5, 79.9 and 97.2%, respectively with CKD while it was 90.6, 84.1, 82.1, 96.8, 93.2, 89.1 and 72.8%, respectively with alum. Results show similarity with that reported in previous studies [39].

In contrast, Fong and Pradeep [40] reported that alum increased zinc level, although it succeeded in reducing lead and iron with removal efficiency reached 99%. They confirmed that this was due to the higher adsorption capacity of the aforementioned parameters, which could be attributed to its nanoscale particle size, which allows access to a larger surface area. On the other hand, for alum, cadmium and iron removal results were agreed with the finding of Taman and Ossman [41].

The obtained results of this study were in agreement with Salem and Sayed [42] who found that, after 3 days of treatment process, CKD removed Ni, Pb, Fe and Zn for 100%. El-Awady and Abo-El-Enein [34] also reported that the removal of Cd and Co increased with an increase of pH up to 8 as he recorded that, removal percent increased from 50% to 90% when pH value increased from 6 to pH 8. He also recorded that, above pH 8, the adsorption was decreased and he thought that was because of an increase in OH^- ions. In contrast, Taha and Dakroury [43] investigated the adsorption of Cd, Co, and Zn using CKD and discovered that the removal of Zn at pH 6.5 was 80%, but increased to 99% at pH 8. For Cd, 99% was removed at pH 6.2.

This ability and performance of CKD for pollutant removal is thought to be due to one of three theories according to previous studies on CKD, [44,45,46]. First, one of them supposed that the main process dominated in CKD application for contaminant removal is pure adsorption which accelerated within certain values of pH [47]. While the second theory rejected the first one and proved that pure precipitation is the responsible mechanism for the treatment [48]. That was proved by increasing the pH to the degree where the hydroxides of the metals were generated. The last theory went on with both previous theories and didn't recognize between the two mechanisms and improve the contribution of adsorption and precipitation mechanisms [49]. Our work in this study also corroborated this third theory.

3.6. Statistical analysis

Significance values have been generated for the mean differences between pairs of the various parameters of the treated water with CKD and Alum and the pretreated water, (DO, BOD, COD, TDS, salinity, conductivity, turbidity, TP, TN and some heavy metals)

There was a significant difference between groups, (Table 2) just as illustrated by the one-way ANOVA Test ($F (2) = 3.95$, $p = 0.025$). A Tukey post hoc test showed that there was a statistically significant difference between the physicochemical parameters of pretreated water and treated water of each CKD and alum ($p = 0.049$ and 0.061). While there was no statistically significant difference between the treatment with CKD and alum ($p = 0.97$). This was similar to what resulted from Scheffe and LSD tests, (Table 3) as they were verification tests. Means for groups in homogeneous subsets also displayed and showed correspondence in the performance of CKD and alum for water treatment statisti-

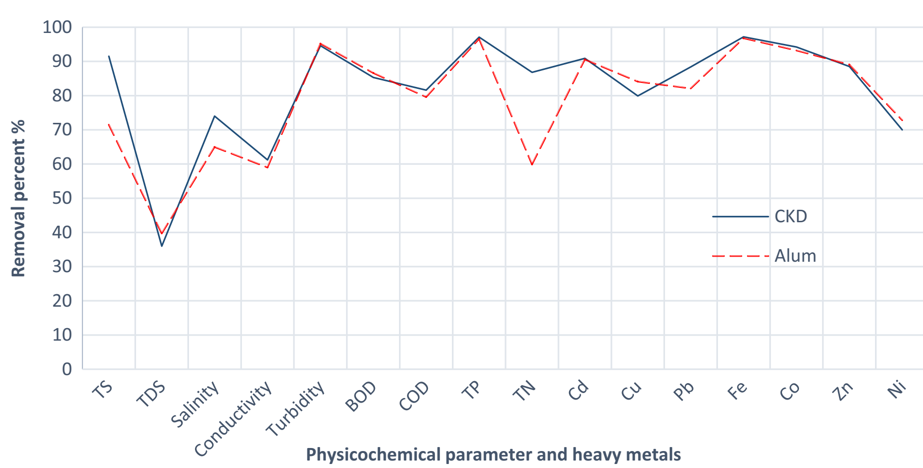

Fig. 4. Treatment with Environmental Alternatives and Alum (Removal percent).

Table 2
Statistical Analysis (ANOVA Test).

	Sum of Squares	df	Mean Square	F	Sig.
Between Groups	1911.650	2	955.825	3.950	0.025
Within Groups	12339.688	51	241.955		
Total	14251.337	53			

Table 3
Post Hoc Tests (Multiple Comparisons).

	(I) VAR1	(J) VAR1	Mean Difference (I-J)	Std. Error	Sig.	95% Confidence Interval	
						Lower Bound	Upper Bound
Tukey HSD	CKD	Alum	−1.029-	5.185	0.979	−13.55-	11.49
		P.W	−13.105-*	5.185	0.038	−25.62-	-0.59-
	Alum	CKD	1.029	5.185	0.979	−11.49-	13.55
		P.W	−12.075-	5.185	0.061	−24.59-	0.44
	P.W	CKD	13.105*	5.185	0.038	0.59	25.62
		Alum	12.075	5.185	0.061	-0.44-	24.59
Scheffe	CKD	Alum	−1.029-	5.185	0.980	−14.10-	12.04
		P.W	−13.105-*	5.185	0.049	−26.18-	-0.03-
	Alum	CKD	1.029	5.185	0.980	−12.04-	14.10
		P.W	−12.075-	5.185	0.076	−25.15-	1.00
	P.W	CKD	13.105*	5.185	0.049	0.03	26.18
		Alum	12.075	5.185	0.076	−1.00-	25.15
LSD	CKD	Alum	−1.029-	5.185	0.843	−11.44-	9.38
		P.W	−13.105-*	5.185	0.015	−23.51-	−2.70-
	Alum	CKD	1.029	5.185	0.843	−9.38-	11.44
		P.W	−12.075-*	5.185	0.024	−22.48-	−1.67-
	P.W	CKD	13.105*	5.185	0.015	2.70	23.51
		Alum	12.075*	5.185	0.024	1.67	22.48

Table 4
Correlations for CKD and Alum.

		P.W	CKD	Alum
P.W	Pearson Correlation	1	0.283	0.364
	Sig. (2-tailed)		0.272	0.151
	N	18	18	18
CKD	Pearson Correlation	0.283	1	0.972**
	Sig. (2-tailed)	0.272		0.000
	N	18	18	18
Alum	Pearson Correlation	0.364	0.972**	1
	Sig. (2-tailed)	0.151	0.000	
	N	18	18	18

**. Correlation is significant at the 0.01 level (2-tailed).

cally. In Table 4, Pearson's correlation coefficient also confirmed this result as CKD and alum treatment results were highly significant (r = 0.972 * *) with p = 0.00 at the 0.01 level (2-tailed).

4. Conclusion

The results signified that CKD under our study proved to be effective for water contaminants and heavy metal removal. It achieved very high closed efficiency when compared to the performance of alum as a coagulant. It achieved removal percent ranged from 61.2 to 97.2% while alum achieved removal percent ranged from 39.7 to 96.8% for all contaminants and after the implementation of the optimum conditions. It also confirmed its ability to improve water quality and enhance its physicochemical parameters. This makes it suitable to be used as a good coagulant and also a good adsorbent according to its characteristics and depending on the results. Thus, instead of being disposed of as solid waste to minimize its effects, the application of CKD in wastewater treatment is an environmental and alternative low-cost technology. Future research into cement kiln dust should examine its removal potential in different types of water, as the water used in this study was raw surface water with simple to moderate contamination. Furthermore, while this experiment measured CKD usage in wastewater treatment, there wasn't any modification method used for improving its performance. Observational studies are required to gain more insight into modification techniques to investigate their usage on a wide scale. Mechanical, chemical, and physical modifications, for example, have been extensively researched in order to improve the removal efficiency of Nano porous adsorbent material.

Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Acknowledgments

The authors would like to thank the Environmental Sciences Department, Faculty of Science - Damietta University in conduct of the research. The authors would also like to thank Suez Cement Company for the supply of the CKD. The comments of the anonymous reviewers are also gratefully acknowledged.

Compliance with Ethical Standards

This paper does not contain any studies with human participants or animals performed by any of the authors.

Consent to participate

Not applicable.

Consent for publication

Not applicable.

Funding

This research did not receive any specific grant from funding agencies in the public, commercial, or not-for-profit sectors.

Author contributions

All authors contributed to the study conception and design. Material preparation, data collection and analysis were performed by **Doaa A. Elemam**, **Amany F. Hasaballah** and **Talaat. A. Hegazy**. The first draft of the manuscript was written by **Mahmoud S. Ibrahim** and all authors commented on previous versions of the manuscript. All authors read and approved the final manuscript.

Data Availability

The datasets generated during and/or analysed during the current study are available from the corresponding author on reasonable request.

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