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**Abstract**

The efficiency of treatment was eliminated based on residual concentration of ammonia-nitrogen ($NH_4$-N) and chemical oxygen demand (COD). Removal percentage of ammonia in case of using $H_2O_2$ increased in a concentration of 30%. the removal of organic pollutants recorded high efficiency of removal in 14.28 and 10.71% for east El-Burullus and Damru drains, respectively. $KMnO_4$ gave better results which, the maximum ammonia removal was 66.59 % for 0.2 g/L of $KMnO_4$ in Hokx site, whereas COD removal was 34.48 % at 0.1of $KMnO_4$ in Hokx site. Acidic conditions (pH 3) demonstrated conducive to the removal of all factors for both oxidants with the exclusion of COD removal in Burullus east that displayed a high removal at pH 5. The reduction of $NH_4$-N at pH 3 for both $KMnO_4$ and $H_2O_2$, with 76.74%, 93.76%, 92.03 % for Burullus east, Damro and Hokx sites, respectively. whereas the higher percentage removal in COD was 23.07% and 8.97% for both Damro and Hokx sites at PH 3, while in Burullus east was 22.22% at pH 5. Furthermore, $Fe^{2+}/H_2O_2$ (Fenton's reagent) was run in the batch process with various doss. Under the optimal parameters, the removal efficiency of COD was achieved 58. 69 % and 12.60 % for both Burullus east and Damro, respectively at $Fe^{2+}$ =0.4 g/L, pH= 3 and contact time of 120 min. The $NH_4$-N and COD removal performance of $KMnO_4$ was well compared with $H_2O_2$. Finally, remediation needn't extra pH adjustment which it is simple operation and low capital cost.

## 1. Introduction

Contamination of soils, sediments, and ground water by toxic organic compounds is a widespread problem. Contaminants of concern usually contain volatile organic compounds for example tetrachloroethene, trichloroethylene, benzene, and semivolatile organic compounds such as phenanthrene, naphthalene and pyrene **(Gates-Anderson *et al.*, 2011)**. Burullus Lake receives wastewater and agricultural drainage water through eight drains. It is contaminated with oil effluent derived from the rerefine process of used lubricating oil and used industrial solvents. As chemical products play an increasingly essential role, a large number of organic contaminants such as polychlorinated biphenyls (PCBs), polycyclic aromatic hydrocarbons (PAHs), pesticides, phthalates total petroleum hydrocarbons (TPH), pharmaceuticals and personal care products (PPCPs), are discharged directly or indirectly into the soil environment **(Sun *et al.*, 2014)**. Although the soil has a certain capacity of adsorption and the environmental conditions allow the degradation of complexes present in the agrochemicals **(Steffen *et al.*, 2011)**, their high toxicity and persistence make necessary an intervention in the area. This is the reason several studies are looking for applying physicochemical techniques for eliminating contaminants, reducing the residual levels to acceptable and safe levels. The remediation of organic-contaminated soil is immediately required to protect public health and the environment. In recent years, advanced oxidation processes (AOPs) have gradually gained attention owing to their efficiency and environmental friendliness for organic-contaminated soil remediation **(USEPA, 2006)** owing to the strong oxidizing probable of hydroxyl radicals (·OH) generated during reaction. The low cost of this process when compared to conventional processes, its rapid, its strong oxidative effect over the great majority of anthropogenic organic compounds, and the remediation possibilities in situ, are the main advantages of this technique over the others **(Chen *et al.* 2001)**. Chemical oxidants convert organic pollutants into carbon dioxide, water and inorganic compounds or less harmful chemicals after direct introduction into the contaminated source. The oxidants used include hydrogen peroxide ($H_2O_2$), persulfate ($S_2O_8^{2-}$), permanganate ($MnO_4^-$), ozone ($O_3$) and Fenton process (OH) **(Siegrist *et al*., 2011).** Potassium permanganate is able to oxidizing organic compound containing aldehyde groups, carbon–carbon double bonds or hydroxyl groups. As an electrophile, permanganate ion is strongly attracted to the electrons in carbon–carbon double bonds found in chlorinated alkenes, borrowing electron density from these bonds to form a bridged, unstable oxygen compound known as hypomanganatediester. This intermediate product further reacts by a number of mechanisms counting hydrolysis, hydroxylation or cleavage. Potassium permanganate extends several advantages such as easy handling, and is a readily soluble solid and highly effective in water and wastewater treatment **(Xu *et al*., 2005)**. In addition, $H_2O_2$ is a strong oxidant and it can be used to generate hydroxyl radicals ($^{\cdot}OH$) with reactivity second only to fluorine. It has been used to oxidize organic matter in industrial wastewaters or domestic for many years **(Ksibi, 2006)**. Therefore, the objectives in this work are to study the efficiency of the hydrogen peroxide and potassium permanganate on the remediation of ammonia and organic material in contaminated soil in laboratory.

## 2. Materials and Methods

### *2.1. Study area*

Lake Burullus is located in Kafr El-Sheikh Governorate, in the northern part of the Nile Delta, Egypt, along the Mediterranean Sea coast and occupies a more or less central position between the two branches (Rosetta and Damietta) of the Nile **(Fig. 1)**. It extends between 31˚ 22̀- 31˚ 26̀ N and 30˚ 33` -31˚ 07̀ E, 70 Km west of Damietta branch and 60 Km east of Rosetta branch, with an area of about 460 km$^2$. It is connected with the Sea by a small outlet (Boughaz), about 50 m wide near EL-Burg village. The length of the lake is about 70 Km, and the width varies between 6 and 17 Km, with surface area of about 70000 fadaan. The depth of the lake ranges between 0.42 and 2.07 m, the eastern sector of the lake is the shallowest, showing an average depth of 0.8 m (**EEAA, 2017**). Three drains were selected containing major activities of Burullus lake. Description of three selected drains was illustrated in **Table 1** according to **Darwish *et al*. (2018)**.

**Table 1:** Description of three selected drains.

| Drain | Description |
|---|---|
| Burullus East | It is located on the western side of the lake and leading to daily input of industrial, agricultural and domestic sewage to the lake.5.8 km in length. |
| Hokx | It is located in the western sector of the lake and leading to daily input of industrial, agricultural and domestic sewage to the lake.16.2 km in length. |
| Damro | It is located on the western side of the lake and leading to daily input of industrial, agricultural and domestic sewage to the lake.18.5 km in length. |

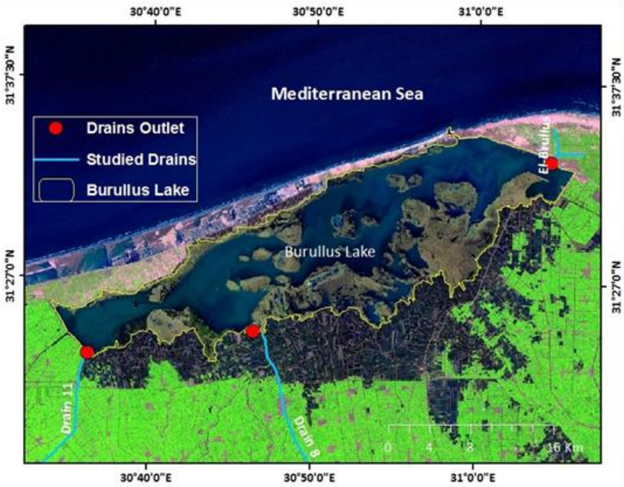


**Figure 1.** Map of Burullus Lake showing the sampling sites.

### *2.2. Sampling and characterization*

The samples used in this study were collected during 2020 year, from three drains of Burullus Lakes called Damro, Hokx and Burullus east. Sediment samples were collected in plastic containers and transported to the laboratory to be stored at room temperature. A total of sampling campaigns were carried out under dry and wet weather conditions to achieve representative sediment samples. Sediments water extract were prepared for remediation processes. Aqueous extracts were prepared at ratio of 1:10 (sediment: water)

### *2.3. Determination of optimum conditions*

In this study, COD was used as an indicator of the amount of organic pollutants in contaminated wastewaters. According to previous studies (**Dehghani *et al*., 2014 &Tony *et al*., 2012)**. Potassium permanganate ($KMnO_4$ 158.03 g/mol) and hydrogen peroxide ($H_2O_2$ 30 %) were used in this study to degradation of specific soil samples containing concentrations of contaminated derivatives, as well as to determine the addition of a greater amount of oxidant increases or decreasing the efficiency in the treatment of soil. For this purpose, prepare soil extract from 100 gm of contaminated sediment sample was added to 250 ml of distilled water in a volumetric flask and the sample was mixed (120 rpm for 60 min) and pH was adjusted to 7. COD and ammoniacal nitrogen were measured and documented as mentioned above. Percentage removal of COD and ammoniacal nitrogen versus concentration of $KMnO_4$ and $H_2O_2$ were plotted and the dosage that gave the maximum removal was considered as the optimum dosage. The effect of pH was studied by varying pH between 3 and 9 and optimum pH was determined at optimal dosages of $KMnO_4$ and $H_2O_2$. To elucidate the role of $Fe^{2+}$ dose in the Fenton process and to determine the most suitable concentration of $Fe^{2+}$, a sequence of trials was conducted at various concentrations of $Fe^{2+}$ from 0.1 to 0.8 g. $H_2O_2$ concentration was 5% and $KMnO_4$ concentration 0.1 g/L, at an initial pH of 3 in room temperature for reaction time 60 min. The removal (%) of $NH_4$ and COD was calculated using the following Eq.1:

$$\text{Removal } (\%) = 100\ (C_0 - C_e)/C_i \quad (1)$$

Where $C_i$ and $C_e$ (mg/L) are the initial concentration and concentration at time *t*, respectively.

## 3. Results and Discussion

### *3.1. Characteristics of sediment*

The characteristics of contaminated water from three selected drains as pH, EC (electrical conductivity), TDS, $NH_3$, $NO_2$, $PO_4$ and Si, were measured and presented in **Table (2)**. In the aquatic systems, pH is an important parameter of the extent pollution in the watershed areas and water quality. **(Jonnalagadda and Mhere, 2001).** The average concentration of pH in water of Hokx, Damro and Burullus East was 8.2, 8.1 and 7.8, respectively. The Electrical conductivity of the study was highest at Burullus east (11.4 µ mho/cm) and lowest in the Hokx Site (3.8 µ mho/cm), while electrical conductivity of Damro site was recorded with (4.2 µ mho/cm). All samples were in desirable limit as prescribed for water standard. Total dissolved solid (TDS), the measure of the material dissolved in sediment suspended in the water, showed slight variations throughout the sampling site. An obvious increase in TDS was recorded at Burullus East (6360 µg/l), while the sampling sites were 1940 µg/l and 2160 µg/l for Hokx and Damro sites, respectively. Nitrate is the final oxidation product of nitrogen compounds in natural oxic water. It was determined using the cadmium reduction column method. The samples were allowed to pass through a glass column containing loosely cadmium filling coated with metallic copper. Nitrates are considered better indicators in average water flow, due to their relatively good solubility and non-reactive behavior. Nitrate ranged between 101.4, 143,9 and 138.8 µg/l for Hokx, Damro and Burullus east sites, respectively. Similar to trend of Ammonia-N, showed slight site-to-site variations with the highest concentration (1419 µm/l) at Hokx site. The lowest concentration (963 µm/l) was however measured at Burullus east, while in Damro site, value of ammonia was 1419 µm/l. Phosphorus and other nutrients motivate the growth of marine plants, counting undesirable algae. Agricultural production systems are one of the many contributors to the phosphorus found in streams and water storages **(Nash and Halliwell, 2000)**. Phosphorus concentrations were normal at the three sampling points which varied from 156.2 nearby Hokx site to 143.9 µg/l nearby Damro site, and137.8 µg/l, in Burullus East (the water of lake nearby sea shore habitat). Silicates varied from 616 µg/l nearby Hokx site to 1146 µg/l close to Damro site in the water of lake shores habitat with a mean value of 1957.88 µg/l, while in Burullus east was 550 µg/l.

**Table 2.** Characteristics of water in three drains of Burullus Lake.

| **Parameter** | **Value** | | |
|---|---|---|---|
| | **Hokx** | **Damro** | **Burullus East** |
| **pH** | 8.2 | 8.1 | 7.8 |
| **EC** (µmho/cm) | 3.8 | 4.2 | 11.4 |
| **TDS** (µm) | 1940 | 2160 | 6360 |
| **$NH_3$** (µm) | 1082 | 1419 µm | 9636 µm |
| **$NO_2$** (µm) | 101.4 | 198.6 | 226.4 |
| **$PO_4$** (µm) | 156.2 | 143.9 | 137.8 |
| **Si** (µm) | 616 | 1146 | 550 |

### *3.2. Optimum dosages of $KMnO_4$ for removal of COD and ammonia*

The results of optimum dosage of $KMnO_4$ determined at different amount (0.1, 0.2, 0.4, 0.8 g/L) and pH value of 8 as presented in Figs. (2&3). It can be seen that $KMnO_4$ was more effective in removing ammonia than COD. The maximum ammonia removal was 66.59 % for 0.2 g/L of $KMnO_4$ in Hokx site, whereas COD removal was 34.48 % at 0.1 g/L of $KMnO_4$ in Hokx site. Excess amount of an oxidant can lead to decrease in removal efficiency as reported by **Wang *et al*. (2000).** While in Burullus east site the percentage removal of ammonia and COD increased with an increase in the amount of oxidant 0.8 and 0.4 g/L with percentage 57.77 and 28.57%, respectively. Potassium permanganate is highly reactive under conditions found in the water. It will oxidize a wide variety of organic and inorganic substances. Potassium permanganate ($Mn^{7+}$) is reduced to manganese dioxide ($MnO_2$) ($Mn^{4+}$), which precipitates out of solution **(CRC,1990)**. The half reaction of potassium permanganate at natural pH is illustrated in Eq. 2.

$$MnO_4^- + 2H_2O + 3e^- \longrightarrow MnO_2 + 4OH^- \quad (2)$$

Potassium permanganate can be partly attributed to remove ammonia concentration from COD, which is a difficult inorganic matter to oxidize as reported by **Vogel *et al.* (2000).** Generally, the amount of COD removed depends on the reaction of organics with inorganics or/and inorganics with inorganics **(Kylefors *et al*., 2003)**. Moreover, it depends on the chemical reaction in raw water, owing to its complex matrix which may result in scavenging hydroxyl radicals. **Kylefors *et al*. (2003)** concluded that Fe(II) and sulphide were the main contributors to inorganic matters that influenced COD reduction.

### *3.3. Optimum dosages of $H_2O_2$ for removal of COD and ammonia*

The removal of COD and $NH_4$ were performed at range 5, 10, 20 &30% dosages of $H_2O_2$ oxidants at pH 8 for 60 min with constant concentration of $KMnO_4$ as shown in Figs. (4&5). The highest removal of ammonia achieved was about 49.05 % at 5 % of $H_2O_2$ in Hokx site. On the other hand, better removal of COD increased with decreased of $H_2O_2$

concentration in Burullus east and Damro with percentage removal 14.28 and 10.71%, respectively. Hydrogen peroxide improved oxygen production and oxidation rate (Eq. 3) (**Chen *et al*. 1996**).

$$2H_2O_2 \longrightarrow 2H_2O + O_2 \quad (3)$$

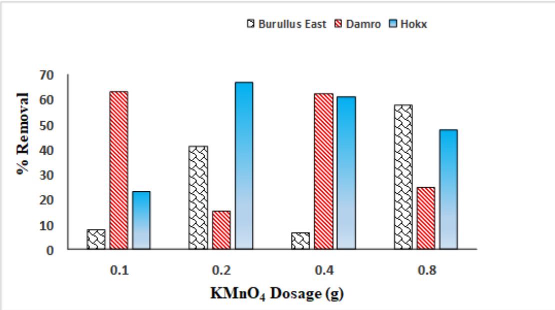


**Fig. 2.** Removal of ammoniacal nitrogen by $KMnO_4$ at different dosages.

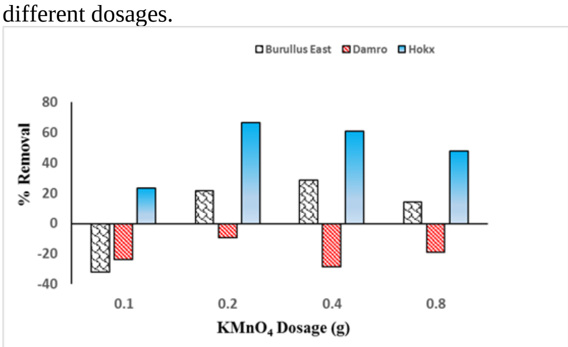


**Fig. 3.** Removal of COD with $KMnO_4$ at different dosages

The percentage of removal of ammonia and COD increased with an increase in the amount of $H_2O_2$ oxidant in Damro and Hokx site. This is can be explained by the increased amount of photons that was absorbed and reacted with the oxidant at high concentration.

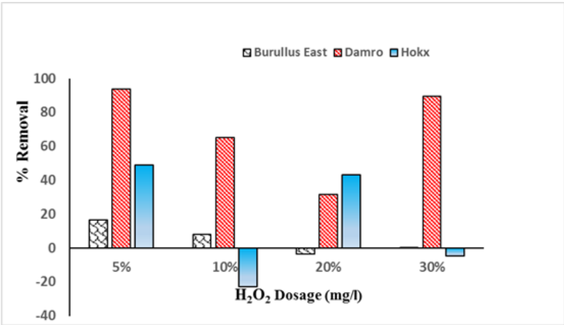


**Fig. 4.** Removal of ammoniacal nitrogen with $H_2O_2$ at different dosages.

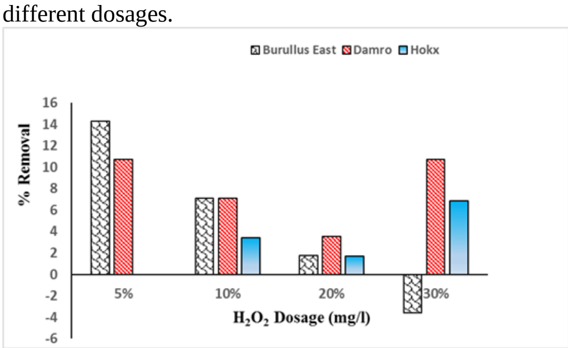


**Fig. 5.** Removal of COD by $H_2O_2$ at various dosages

***3.4. Determination of optimum pH for removal of COD and ammonia***

Determination of optimum pH is generally required to be established to achieve maximum removal efficiency. An optimum pH can be achieved at a fixed dosage of $KMnO_4$ on 200 mg/L, whereas the dosage of $H_2O_2$ was a fixed at 5 %. The range of pH of samples was kept at 3, 5, 7 & 9 at fixed contact time of 60 min. Figure (6) shows the removal of COD at various pH values for both oxidants in different sampling sites. Most of the organic matter was eliminated under acidic condition for both the oxidants and the highest removal of 22.22 %, 23.08 % and 8.97 % were obtained at pH 5, 3 and 3 in Burullus east site, Hokx site and Damro site, respectively. In the $H_2O_2$ system, the dissolved iron ions in sediment $Fe^{2+}$ reacted with $H_2O_2$, resulting in Fenton reaction and formation of hydroxyl radicals ($^{-}OH$) (Eq. 4).

$$Fe^{2+} + H_2O_2 \longrightarrow Fe^{3+} + OH^{-} + OH^{\cdot} \quad (4)$$

**Hermosilla *et al*. (2009)** $^{-}OH$ has the possible to destroy organic pollutants, while the optimum pH value for Fenton reaction ranged between 2.5 and 4. Generally, the oxidation potential of $KMnO_4$ (Eq. 5) in acidic medium ($E_0$ = 1.68 V) is higher than that in alkaline medium (E0 = 0.60 V) (**CRC 1990**).

$$MnO^{-}_4 + 4H^{+} + 3e^{-} \longrightarrow MnO_2 + 2H_2O \quad (5)$$

Higher removal of $H_2O_2$ under acidic conditions is in agreement with **Lim *et al*. (1997),** who concluded that the removal of organic matter in water is influenced by the pH of water. Figure (7) shows the effect of pH on ammoniacal nitrogen removal. Oxidation process using $KMnO_4$ and $H_2O_2$ seems to be more effective in removing ammoniacal nitrogen under acidic conditions, especially at pH 3, with highest percentage removal was 76.74%, 93.79 % and 92.03% for Burullus east, Hokx and Damro sites, respectively. The results showed that the percentage removal decrease when pH increased the sediment content, which can be attributed to better oxidation of the organic matter. $KMnO_4$ is an effective oxidant of manganese and the change in pH can be attributed to the precipitation of manganese and its different forms depending on the pH, which, the manganous ($Mn^{2+}$) form is oxidized to manganic ($Mn^{4+}$). **Trebouet *et al*. (2001)** mention that at low pH, phenol and carboxylic functional group from humic matter are protonized and decreased the charges of humic matter. It is reported that permanganate is effective in the remediation of many pollutants, which demands a pH for the soil between 3 and 10 **(Ferrarese *et al*. 2008).** Despite the relatively low standard oxidation potential, permanganate salts are considered strong oxidizing agents, able to break organic molecules containing carbon– carbon double bonds, aldehyde groups, and hydroxyl groups **(Brown *et al*., 2003).** According to the theory of Fenton reaction, an increase in pH not only inhibits the production of OH· radical, but also causes the precipitation of $Fe^{2+}$ in the solution which loses the catalytic ability of $Fe^{2+}$. Generally, Fenton reaction has a high degradation efficiency at an initial pH of 3 in wastewater treatment. Based on Fenton reaction for wastewater remediation, it is vital to conduct the pre-acidification treatment to reduce the initial pH value to 3 in order to improve the Fenton reaction.

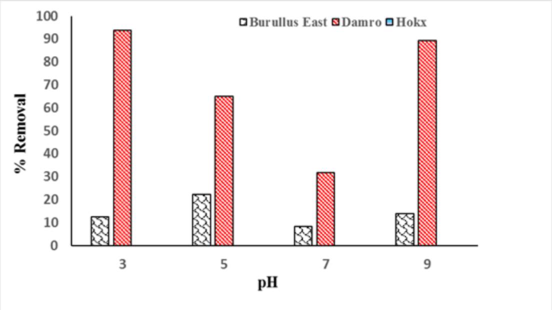


**Fig. 6.** Effect of pH values on removal of COD by using $H_2O_2$ and $KMnO_4$.

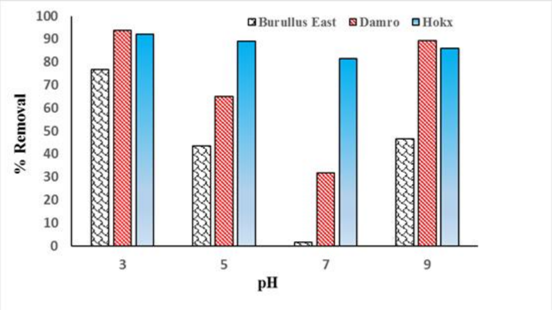


**Fig. 7.** Effect of pH values on removal of ammoniacal nitrogen by using $H_2O_2$ and $KMnO_4$.

### *3.5. The effects of Fenton ($Fe/H_2O_2$) ions concentration for removal of COD*

In the first experiment, the optimum initial $Fe^{2+}$ ion concentration was determined at pH 5 and 30 °C. The results in **Fig. (8)**, showed that the optimum $Fe^{2+}$ concentration was found to be 0.4 g/L, with percentage 58.69%, 12.60 % and 7.81% COD removal for Burullus east, Damro sites and Hokx sites, respectively after 60 min of contact time. The minimum results of the COD removal at the adjusted conditions, showed at 0.8 g/L, from $Fe^{2+}$ dose, while the concentration of $Fe^{2+}$ has a negative effect on 0.1 g/L, of the reaction kinetics due to $Fe^{2+}$ ions reacts with $OH^{\bullet}$ and deactivates them well and can inhibit the reaction rate **(Ebrahiem *et al*., 2017).** The $^{-}OH$ radicals are trapped through $Fe^{2+}$ ions in extra as exposed in the reaction Eq. (6) **(Bouasla *et al*., 2010).**

$$Fe^{2+} + OH^{\bullet} \longrightarrow Fe^{3+} + {}^{\bullet}OH \quad (6)$$

Moreover, the concentration of $Fe^{2+}$ should be low because economic and environmental reasons **(Ramirez *et al*., 2007).** Also, these observations conform to the results of **Safa and Mehrasbi (2019) and Kositzi (2004).**

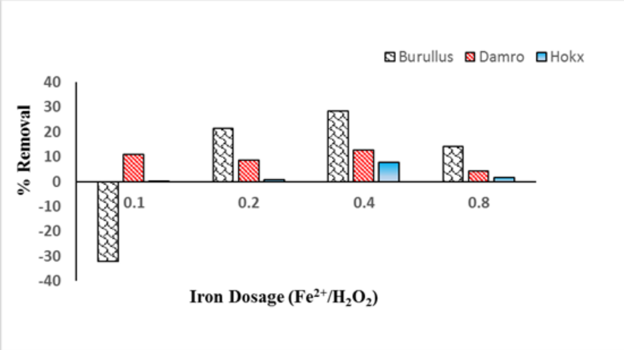


**Fig. 8.** Fenton ($Fe^{2+}/H_2O_2$) ions concentration for removal of COD.

## 4. Conclusion

In this study, the Fenton method and permanganate were selected as chemical and rapid methods for remediation of extracted water from three contaminated sediments, Burullus lake, Egypt. was applied to remove the organic matter at room temperature. The effects of operational parameters in the Fenton method and permanganate on the removal of organic matter were studied. The following results were obtained:

**1.** At the optimum dosage conditions and effect of $H_2O_2$ dosage was moderate removal of ammoniacal nitrogen (16.38 and 49.05 %) at 5% in Burullus east and Hokx, respectively, while the higher removal of Damro drain was 24.45% at 30% of concentration of $H_2O_2$.

**2.** The removal of COD was considerably lower (14.28 %) compared with ammoniacal nitrogen. The removal performance improved for $KMnO_4$ at acidic pH values, which the removal of ammoniacal nitrogen was (57.77, 63.10 and 66.59 %) at 0.8g, 0.1g and 0.2g of $KMnO_4$ dosage in Burullus east, Damro and Hokx sites, respectively. however, 0.4g of $KMnO_4$ dosage provide the higher percentage removal (28.57 and 17.24%) in both Damro and Hokx sites, respectively.

**3.** Consistent with the results the COD oxidation by the Fenton process has moderate removal 58.69% with dosage of Fe ion 0.4 g in Burullus east, while in Damro and Hokx sites have been little removal.

**4.** The removal of ammoniacal nitrogen and COD was better acidic pH 3 in three sampling sites by two oxidants. Exception, the removal of COD was maximum at pH 5 for Burullus East site.

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