

## Algal control and enhanced removal in drinking waters in Cairo, Egypt

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### ABSTRACT

Algal blooms at the major water treatment plants in Egypt have been reported since 2006. While previous studies focused on algal types and their correlation with disinfection by-products, correlation between raw water quality and algal blooms were not explored. Therefore, a survey of Nile water quality parameters at a major water intake in the Greater Cairo Urban Region was conducted from December 2011 to November 2012. Bench-scale experiments were conducted to evaluate the effectiveness of the conventional chloride/alum treatment compared with combined Cl/permanganate pre-oxidation with Al and Fe coagulants during the outbreak period. Addition of permanganate (0.5 mg/L) significantly reduced the chlorine demand from 5.5 to 2.7 mg/L. The applied alum coagulant dose was slightly reduced while residual Al was reduced by 27% and the algal count by 50% in the final treated waters. Applying ferric chloride and ferric sulfate as coagulants to waters treated with the combined pre-oxidation procedure effectively reduced algal count by 60% and better the total organic carbon reduction and residual aluminum in the treated water. Multivariate statistical analysis was used to identify the relationship between water quality parameters and occurrence of algae and to explain the impact of coagulants on the final water quality.

**Key words** | algal control, multivariate statistical analysis (MVS), permanganate/chloride pre-oxidation

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### INTRODUCTION

In Egypt, the majority of the population depends upon the river Nile as the main source for drinking water and for industrial and agricultural activities. However, after the completion of the Aswan High Dam in 1966, stream-flow regulation has caused significant changes to the Nile aquatic ecosystem and waterway environment (Ismail & Ramadan 1995; El-Shinnawy *et al.* 2000; Talling *et al.* 2009). This change in flow led to the retention of water masses in reservoirs behind dams and barrages in some regions within the Nile system, which provided favorable conditions and ample time for phytoplankton development. Subsequently, this altered the aquatic vegetation which is a source of nutrients for phytoplankton and algal utilization.

Since 2006, specifically in September/October, incidents of algal blooms involving the major water treatment

plants all over the country have been reported by the Egyptian media. The increase in algal growth was attributed to the extended storage time of raw water on site before treatment as well as the lack of regular maintenance and pipe cleansing, an event scheduled every 6 months (Al Ahram Newspaper, 28 September 2006). Also, the prolonged residence time of water masses in the distribution system was reported to promote unexpected algal growth and microbial re-growth. Eventually, this caused an increase in algal toxins within the water distribution system during this period which was reflected by reports of at least 85 sick and one documented death in 2006 (Egyptian Minister of Health, 15 October 2006).

Generally, algae are widely dispersed throughout lakes and rivers at relatively low-population densities; however,

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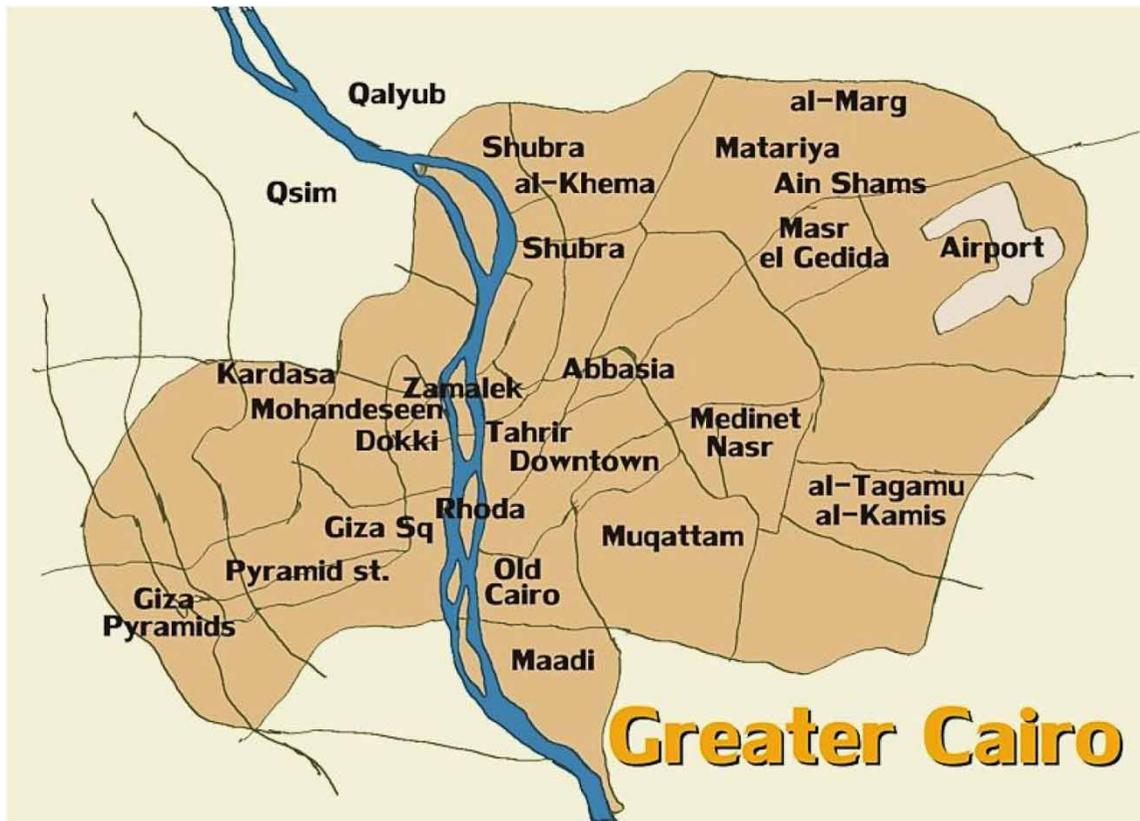
the population is liable to rapidly grow with the increase in nutrients in surface waters (Henderson *et al.* 2008). This may result in dissolved oxygen depletion, high water turbidity and eventually the degradation of aquatic ecosystems (Manahan 2000; Abou El-Ella *et al.* 2007; Shen *et al.* 2011; Wang *et al.* 2013). Also, this may cause acute disturbance in taste, odor and induce toxins under stressful environmental conditions (Shehata *et al.* 2008; Heng *et al.* 2009; Shen *et al.* 2011). In a conventional water treatment plant, uncontrolled algal blooms may clog screens and rapid sand filters and hinder the drinking water supply system (Babel & Takizawa 2011; Wu *et al.* 2011). Alternatively, due to their microscopic size, algae may penetrate filters and reside within the domestic drinking water supply storage systems and excrete extracellular organic products that may lead to an increase in the coagulant demand, impair the final water disinfection process and promote microbial re-growth in the water distribution systems (Ma & Liu 2002; Babel & Takizawa 2011; Wu *et al.* 2011). Therefore, reduction of surface water algae during the initial stages of treatment is preferred to maintain acceptable final water quality (James & Conrad 2004; Henderson *et al.* 2008; Babel & Takizawa 2011; Wu *et al.* 2011). This is mainly because the removal of algal cells as suspended solids may reduce the associated soluble toxins that may impair the physico-chemical water treatment systems.

The general techniques applied for algal control in drinking water treatment systems have been discussed elsewhere (Henderson *et al.* 2008; Gao *et al.* 2010; Babel & Takizawa 2011; Shen *et al.* 2011; Wu *et al.* 2011; Devrimci *et al.* 2012). Of these operations, coagulation/flocculation is regarded as the main process for removal of algae as it has the ability to remove suspended solids through particle destabilization while flocculation allows for solid separation either through settling or flotation, and filtration (James & Conrad 2004; Henderson *et al.* 2008; Gao *et al.* 2010; Shen *et al.* 2011; Devrimci *et al.* 2012). However, due to their small cell size and low specific gravity, removal of algae may not be effectively achieved through using chemical coagulation alone (Devrimci *et al.* 2012). Therefore, chemical pre-oxidants (such as ozone, chlorine dioxide, chlorine, or permanganate) are required to enhance coagulation as they have the capacity to inactivate algal cells (Ma & Liu 2002; Chen & Yeh

2005; Henderson *et al.* 2008; Shehata *et al.* 2008; Heng *et al.* 2009; Gao *et al.* 2010). Overall, chlorine is the preferred pre-oxidant because it is a powerful water disinfectant that is safer to apply, is more economical than other algicides and is less time-consuming in application (Shen *et al.* 2011). Nevertheless, it was reported that algae may consume large quantities of chlorine and thus reduce the free chlorine available to control bacteria if they persist after treatment (Shen *et al.* 2011). On the other hand, it was reported that overdosing of pre-oxidants must be avoided as it can cause algal cell lysis and release undesirable toxins or offensive taste- and odor-associated compounds (Henderson *et al.* 2008; Heng *et al.* 2009).

In Egypt, a typical drinking water treatment system comprises stages of pre-chlorination, flocculation/coagulation with alum, filtration (14 sand and carbon filters), post-chlorination, and settling in distribution tanks (Donia 2007). In this set-up, pre-chlorination is regarded as the most important stage for the control of bacteria and algal growth (El-Dib & Rizka 1995; Donia 2007; Badawy *et al.* 2012); therefore, a number of studies were conducted to investigate the contribution of this stage toward the formation of disinfection by-products in the final treated waters and to set recommendations for its application (El-Dib & Rizka 1995; Donia 2007; Badawy *et al.* 2012). Nonetheless, in a review by Donia (2007), it was suggested that there was a need to upgrade the current treatment technologies in Egypt in order to efficiently handle the recent added loads of chemical and biological pollutants transported through surface and drinking water systems. However, there has been no investigation exploring the relationship between the raw water quality parameters and the reported algal blooms. Also, no study has been conducted to evaluate the impact of the current applied treatment procedures upon the final treated water parameters in case a combined pre-oxidation stage was applied.

Thus, the aim of the current study was to survey the water quality parameters of Nile surface water at El-Maadi, Greater Cairo Urban Region (Figure 1) during the period December 2011 to November 2012. Multivariate statistical analysis was used to identify the contribution of the measured parameters toward occurrence of algal blooms. In addition, an investigation of methods to improve the



**Figure 1** | Map of greater Cairo urban region indicating the location of sampling site along the River Nile in El-Maadi district (▲).

efficiency of the current applied pre-oxidation/coagulation procedure through use of a combined pre-oxidation (permanganate and chlorine) and the use of the different coagulants (alum, ferric chloride and ferric sulfate) was undertaken during the algae outbreak period. Finally, the effect of this combined pretreatment/coagulation upon the final main water parameters and the residual metal (aluminum/iron) concentration in the treated water was assessed using multivariate statistical analysis.

## MATERIALS AND METHODS

### Sampling site

The sampling site is located at El-Maadi district (Figure 1), south of the Greater Cairo Urban Region, Egypt (latitude:  $29^{\circ}56'53.3''$ N; longitude:  $31^{\circ}15'24.6''$ E). This location represents a major water intake point for a number of water

treatment plants operating south of the capital city. Generally, the intake pipe for a water treatment plant is neither totally immersed at the bottom of the Nile nor set on the surface to avoid the intake of silt and other floating pollutants, respectively. It is common for the water treatment intake pipes for these plants to be situated at an ample distance upstream from the urban district which it is to serve. However, due to the random urbanization along the river banks, the sampling location was found to be bordered to the north and east by mixed agricultural and residential areas, and to the west and south there are vast agricultural lands.

### Physical and chemical analysis of water

Water samples (5 L) were collected four times per month from December 2011 to November 2012. Temperature and pH of the samples were measured on site. The samples

were transferred immediately to the laboratory for physical and chemical characterization according to *Standard Methods* (APHA 2005). The parameters of interest were measured by the following methods: turbidity: nephelometry; total dissolved solids (TDS): electrical conductivity; total alkalinity: titration (pH 4.5);  $[\text{Ca}^{2+}]$  and  $[\text{Mg}^{2+}]$ :  $\text{Na}_2\text{EDTA}$  titration; chlorides: argentometric method;  $(\text{SO}_4^{2-})$ : turbidometry; barium chloride: spectrophotometry;  $\text{PO}_4^{3-}$ : stannous chloride colorimetry; total organic carbon (TOC): persulfate-ultraviolet or heated-persulfate oxidation; silicates: silicomolybdate colorimetry. Nitrates were determined using the sodium salicylate method for nitrate determination in drinking waters (Scheiner 1974). Algal count was determined using a Sedgwick–Rafter counting chamber (APHA 2005). Residual content of Al, Fe and Mn in the final treated waters was determined by Eriochrome Cyanine R; phenanthroline, and persulfate methods, respectively (APHA 2005). An average of 4–5 determinations per parameter was undertaken, totaling 16–20 times per month.

### Jar test experiments: pre-oxidation and settling tests

Jar tests were conducted to evaluate algal removal by pre-oxidation followed by coagulation using a six-paddle stirrer (JLT 6, VELP Scientifica, Italy) in surface water samples at room temperature (about 20 °C). This method was used to determine the optimal oxidant as well as the coagulant demand/dose (APHA 2005).

### Statistical analysis of data

Cluster analysis was performed using Ward's method for minimum variance and the similarity coefficient used was the square Euclidean distance function using 'StatistiXL 1.8' within Microsoft Excel 2007 (Microsoft® Windows 2007). Hierarchical clustering was performed for the water chemical and physical characteristics before and after treatment and the results obtained included the distance matrix, clustering strategy report and dendrogram were utilized in data interpretation accordingly.

## RESULTS AND DISCUSSION

### Nile water quality and algal types present in raw Nile waters

Table 1 shows the mean monthly chemical and physical water quality parameters obtained from December 2011 to November 2012. The data showed that a higher algal count was obtained during September to November 2012. This increase may be attributed to agricultural wastewater discharge and the accumulation of pollutants and nutrients south of the sampling point (Amer & Abd El-Gawad 2012). The TDS ranged from 221 to 314 mg/L with the highest value in winter and the lowest in June 2012, which is similar to that reported by Amer & Abd El-Gawad (2012) near the study site in 2010. Generally, it has been reported that the salinity of Nile water increased going from Aswan High Dam (160 mg/L) to Cairo (around 260 mg/L) (Wahaab & Badawy 2004; Talling 2009). This was attributed to the release of agricultural wastewaters containing excess chemicals along the river. In addition, it has been reported that higher TDS values recorded during winter for the Nile waters were due to the dam closure period, during which the amount of water released from the behind the dam was less than that in summer. On the other hand, a decrease in TDS within the Nile system was related to the increase in suspended particles during the flooding period as these particles may adsorb the dissolved solids and subsequently reduce the TDS by sedimentation (Abd El-Hady 2014). The data also revealed that the highest water turbidity was observed during the winter while the lowest was during the summer, an observation that was attributed to the winter closure period (Emara *et al.* 2012). High turbidity of the Nile stream was attributed to the increase in water nutrient content (Amer & Abd El-Gawad 2012), which is in agreement with the current findings.

Nile water temperatures varies from 13 °C in winter to ≈30 °C in summer (Talling 2009; Abd El-Hady 2014). This variation in temperature was reported to be an indicator of water transparency which had a direct impact upon regulating algal/phytoplankton population in these waters (Abd El-Hady 2014). In addition, higher temperatures accelerate the

**Table 1** | Monthly mean variation of Nile raw water physical and chemical parameters (December 2011 to November 2012)

Month	Algae (unit/mL)	Turbidity (NTU)	Temp (C)	pH	TDS (mg/L)	Silica (mg SiO <sub>2</sub> /L)	TOC (mg/L)	Total alkalinity (mg CaCO <sub>3</sub> /L)	Ca <sup>++</sup> (mg/L)	Mg <sup>++</sup> (mg/L)	Cl <sup>-</sup> (mg/L)	SO <sub>4</sub> <sup>2-</sup> (mg/L)	PO <sub>4</sub> <sup>3-</sup> (µg/L)	NO <sub>3</sub> <sup>-</sup> (×10 <sup>-2</sup> ) (mg N/L)
December	5,200 ± 1,112	6.3 ± 1.6	15.3 ± 0.7	7.9 ± 0.1	314 ± 20	1.58 ± 0.1	3.84 ± 0.2	162 ± 4.32	32.0 ± 1.2	18.0 ± 0.2	36.0 ± 2.2	40.0 ± 2.0	25.0 ± 4.0	7.0 ± 0.01
January	4,350 ± 302	6.0 ± 0.8	13.4 ± 0.3	7.9 ± 0.1	288 ± 9	1.96 ± 0.2	4.52 ± 0.47	144 ± 5.2	38.0 ± 1.8	19.5 ± 0.96	37.2 ± 1.3	40.6 ± 2.7	8.0 ± 1.8	5.0 ± 1.4
February	3,300 ± 362	4.6 ± 0.7	14.0 ± 0.4	7.9 ± 0.1	268 ± 13	2.63 ± 0.4	4.64 ± 0.20	134 ± 1.6	34.4 ± 1.6	17.0 ± 1.6	32.0 ± 3.7	42.0 ± 2.7	7.3 ± 0.5	3.0 ± 0.8
March	4,000 ± 307	4.3 ± 0.2	16.30 ± 1.0	8.2 ± 0.1	254 ± 1.63	3.67 ± 0.4	5.69 ± 0.3	140 ± 1.6	30.0 ± 1.5	15.8 ± 0.5	27.7 ± 0.8	33.0 ± 2.6	9.0 ± 1.4	3.7 ± 0.8
April	3,800 ± 204	4.4 ± 0.3	22.00 ± 1.6	8.3 ± 0.1	253 ± 1.2	2.56 ± 0.4	4.89 ± 0.4	141 ± 1.2	29.5 ± 1.0	15.0 ± 0.2	28.0 ± 0.9	32.0 ± 1.4	10.0 ± 1.4	3.50 ± 0.8
May	3,100 ± 378	4.9 ± 0.4	25.70 ± 0.5	8.2 ± 0.1	240 ± 8.8	3.33 ± 0.5	6.99 ± 0.4	142 ± 1.6	28.0 ± 0.3	14.3 ± 0.7	24.5 ± 1.1	29.8 ± 1.5	8.0 ± 1.8	5.00 ± 0.8
June	2,350 ± 270	4.5 ± 0.4	27.00 ± 0.5	7.9 ± 0.1	221 ± 3.5	3.74 ± 0.1	5.36 ± 0.9	118 ± 15.1	26.0 ± 0.2	12.0 ± 0.9	20.0 ± 0.7	23.8 ± 2.7	6.0 ± 0.8	2.40 ± 0.8
July	2,500 ± 264	3.4 ± 0.3	29.20 ± 0.5	7.9 ± 0.0	223 ± 2.2	4.88 ± 0.5	4.30 ± 0.2	124 ± 3.7	25.7 ± 0.8	10.2 ± 0.4	19.7 ± 0.2	24.0 ± 0.5	5.0 ± 0.8	2.70 ± 0.8
August	3,150 ± 508	4.2 ± 0.3	29.70 ± 0.9	7.8 ± 0.0	230 ± 3.7	4.03 ± 0.1	3.95 ± 0.2	136 ± 3.65	24.0 ± 0.2	9.6 ± 0.8	21.2 ± 0.8	25.7 ± 0.8	9.0 ± 3.2	4.00 ± 1.6
September	6,500 ± 749	6.5 ± 0.8	27.70 ± 0.2	8.0 ± 0.1	257 ± 3.7	4.4 ± 0.1	4.37 ± 0.2	142 ± 1.6	26.4 ± 0.8	11.2 ± 0.3	23.5 ±	28.6 ± 0.7	16.0 ± 1.6	7.00 ± 1.4
October	7,100 ± 460	6.7 ± 0.3	25.50 ± 0.5	8.0 ± 0.1	269 ± 3.7	2.09 ± 0.8	3.76 ± 0.1	152 ± 5.2	27.0 ± 0.2	14.6 ± 0.5	27.5 ± 1.5	39.6 ± 4.4	17.0 ± 1.6	7.50 ± 1.6
November	8,300 ± 122	6.4 ± 0.2	22.20 ± 2.0	8.1 ± 0.1	277 ± 2.2	2.6 ± 0.1	4.74 ± 0.9	154 ± 1.6	30.0 ± 0.5	17.3 ± 0.4	31.0 ± 1.3	36.4 ± 2.0	20.0 ± 2.5	9.00 ± 1.4

The monthly data represents the mean value of 4 weeks determinations averaging (4 × 4) or (4 × 5) replicates.

metabolic rate of microalgae (Shen *et al.* 2011). The presence of carbonates in these waters was reported to be dependent upon both the pH and salinity (TDS) (El-Ghandour *et al.* 1985), and the increase in alkalinity (CaCO<sub>3</sub>) of these raw waters has been reported to promote phytoplankton growth (Abd El-Hady 2014) as a result of algal utilization of dissolved CO<sub>2</sub> and HCO<sub>3</sub> in biomass formation (Manahan 2000).

Overall, it was observed that there was an increase in Ca, Mg, SO<sub>4</sub>, Cl, PO<sub>4</sub> and NO<sub>3</sub> content during the winter period, in accordance with the findings of Emara *et al.* (2012). The phosphate and nitrate contents of these waters showed a similar pattern of increase and decrease over the study period. This may be attributed to the discharge of agricultural drainage (El-Ghandour *et al.* 1985; Ismail & Ramadan 1995; Wahaab & Badawy 2004) as well as from surrounding industrial and urban sources (Elewa 2010). Finally, the silica content of these waters ranged between 1.58 and 4.88 mg SiO<sub>2</sub>/L with the highest in July and the lowest in December. Shehata *et al.* (2008) reported that low silica levels were associated with the increase in diatoms present in these waters. This variation in silicates was also attributed to the fact that diatoms utilized dissolved silicates for the formation of skeletal materials during algal productivity (Brownlee & Taylor 2002).

Diatoms predominated over Chlorophyta, Cyanophyta and Euglenophyta (Table 2). This agrees with the findings of Abd El-Hady (2014) who showed that phytoplankton in the Nile system were dominated by Bacillariophyta (diatoms) all year round. In addition, the prevalence of diatoms within the river Nile was attributed to the presence of iron and dissolved silica (Shehata *et al.* 2008). On the other hand, while the predominance of diatoms is regarded as an indicator of good water quality under low stress conditions, the increase in Chlorophyceae and Cyanophyceae is considered an indication of organic pollution and nutrient accumulation in surface waters (Mahapatra *et al.* 2010).

### Multivariate statistical analysis of raw water data and its correlation with algal types

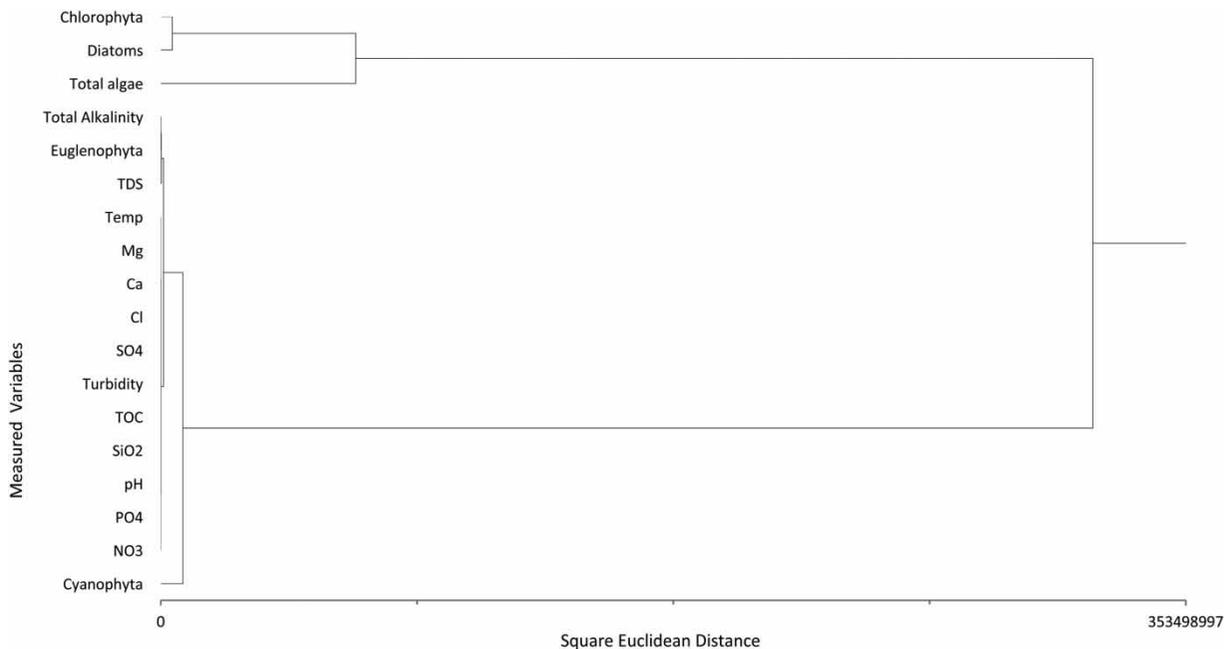
Generally, it has been reported that the important factors controlling algal growth are light, temperature, the chemical composition of water and its acidity or alkalinity

**Table 2** | Mean monthly occurrence of total algae and different types of algae in Nile raw water (December 2011 to November 2012)

Month	Total algal count (unit/ml)	Diatoms	Diatoms (%)	Chlorophyta	Chlorophyta (%)	Cyanophyta	Cyanophyta (%)	Euglenophyta	Euglenophyta (%)
December	5,200	2,700 ± 454	51.9	1,850 ± 500	35.6	550 ± 129	10.6	100 ± 41	2
January	4,350	2,300 ± 216	52.9	1,400 ± 82	32.2	500 ± 0.0	11.5	150 ± 41	3.4
February	3,300	1,750 ± 208	53	1,000 ± 163	30.3	400 ± 82	12.1	150 ± 41	4.7
March	4,000	1,980 ± 107	49	1,400 ± 163	35	500 ± 82	12.5	100 ± 41	3.5
April	3,800	1,900 ± 81	50	1,100 ± 141	28.9	700 ± 82	18.4	100 ± 41	2.7
May	3,100	1,500 ± 294	48.4	1,050 ± 58	33.9	500 ± 82	16.1	50 ± 0.0	1.6
June	2,350	1,150 ± 173	48.9	800 ± 141	34	350 ± 58	14.9	50 ± 0.0	2.2
July	2,500	1,100 ± 82	44	850 ± 129	34	500 ± 82	20	50 ± 0.0	2
August	3,150	1,400 ± 282	44.4	1,150 ± 129	36.5	700 ± 82	22.2	100 ± 41	2.5
September	6,300	2,900 ± 529	46	2,450 ± 129	38.9	1,000 ± 163	15.9	150 ± 41	2.5
October	7,100	3,750 ± 342	52.8	2,650 ± 129	37.3	650 ± 129	9.2	150 ± 41	0.8
November	8,300	4,300 ± 115	51.8	3,150 ± 129	37.9	700 ± 82	8.4	150 ± 41	1.8

(Shen *et al.* 2011). However, to better explain the dependency of the algal content of these waters upon the measured parameters, hierarchical cluster analysis was performed using the analytical data obtained for the raw waters and the identified types of algae present and the result is depicted in a dendrogram (Figure 2). The

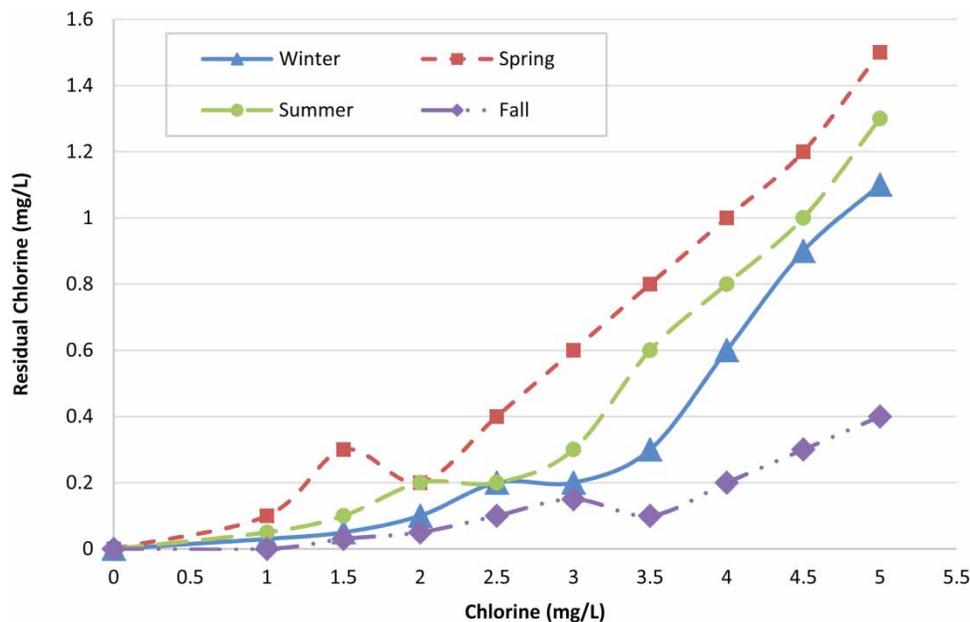
dendrogram indicated the split of the data into 17 clusters that were grouped into two main groups. The first of the multi-cluster groups indicated that the Euglenophyta content was strongly dependent upon the total alkalinity ( $\text{CaCO}_3$ ) and the TDS of these waters. This may be attributed to the fact that Euglenophyta may utilize these

**Figure 2** | Dendrogram depicting hierarchical clustering and distance between the mean monthly Nile water chemical and physical variables and type of algae using Ward's methods for minimum variance.

components as nutrients during growth (Nweze & Ude 2013). Cyanophyta also showed a strong dependency upon all chemical parameters measured as well as on Euglenophyta in these waters. This may be attributed to the fact that Cyanophyta is a photosynthetic nitrogen-fixing group that thrives upon the increase in nutrient content (Nweze & Ude 2013). The second group indicated that both the Chlorophyta and diatoms contents were the major contributors toward the measured total algal count. Even though this was attributed to the pH of raw waters, the abundance of these types of algae may be associated with the increase in nutrients (Nweze & Ude 2013). Overall, the data revealed that these two groups of algae were also affected by the presence of Euglenophyta and the water TDS. There is significant correlation shown between phosphates and nitrates contents as well as for the turbidity, TOC and SiO<sub>2</sub> content, both of which were found to be affected by the pH of these waters. This may signify the role that these components play in the formation of algae and their growth as their abundance was related to slightly alkaline pH waters (Brownlee & Taylor 2002). On the other hand, it was reported that the significant correlation between nitrate and phosphate may be indicative of their origin, i.e., agricultural sources (Nweze & Ude 2013).

### Evaluating the efficiency of the chlorine/alum water treatment process upon algal removal and residual turbidity

Overall, the efficiency of the pre-chlorination stage is governed by two indicators, i.e., the chlorine demand and residual chlorine content. The chlorine demand determines the chlorine dose applied during the pre-chlorination/disinfection process and the residual chlorine signifies that sufficient amount of chlorine has been added to inactivate bacteria and some viruses that may cause diarrheal disease in drinking water (WHO 2011). Technically, a minimum of 0.5 mg/L residual chlorine must be maintained to ensure water protection from recontamination during storage and that residual chlorine levels between 0.2 and 0.5 mg/L at the consumer outlet water given the extremities of the supply network (WHO 2011). Figure 3 shows the seasonal variation in chlorine oxidant demand and the corresponding residual chlorine. It was noted that as the algal count increased from 3,000 to 7,000 units/mL (spring and winter), the chlorine dose breakpoint increased from 2.0 to 3.5 mg/L. As well, in order to achieve the required residual chlorine of 0.2–0.5 mg/L, the optimal chlorine dose applied was 3.7 mg/L, 2.7 mg/L, 3.3 mg/L and

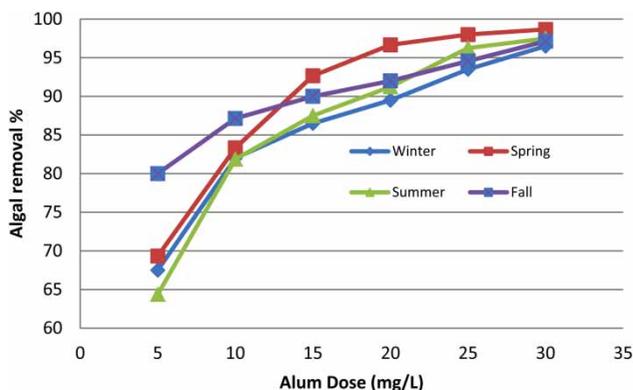


**Figure 3** | Seasonal variation in chlorine demand and residual chlorine for mean seasonal algal counts: winter (4,000 units/mL) spring (3,000 units/mL), summer (3,200 units/mL) and fall (7,000 units/mL) (December 2011–November 2012).

5.5 mg/L for the winter, spring, summer and fall algal count, respectively.

Generally, chlorine disinfection effectivity depends on the applied dose and contact time (Casey *et al.* 2012). Previous work by El-Dib & Rizka (1995) recommended, as a precaution, to monitor the applied dose and not to exceed the chlorine contact time over 30 minutes for Nile waters. This was based upon their findings that the concentration of trihalomethanes (THMs) formed increased progressively as the contact time extended from 30 to 240 minutes. In addition, low chlorine doses may readily aid in the formation of chloro-substituted by-products, while higher chlorine doses may lead to production of oxidation and cleavage products of natural organic matter (NOM) and formation of THMs.

The hydrated form of  $\text{Al}_2(\text{SO}_4)_3$  ( $\text{Al}_2(\text{SO}_4)_3 \cdot n\text{H}_2\text{O}$ , where  $n = 14\text{--}18$ ) is widely used as a coagulant in drinking water treatment (Matilainen *et al.* 2010; Babel & Takizawa 2011; Wang *et al.* 2011; Noblet 2012). Use of alum is beneficial as it does not damage algal cells (Babel & Takizawa 2011) and also removes a large portion of the NOM that has a slower reaction rate with chlorine (Gang *et al.* 2003). Technically, it was reported that a typical alum dose between 5 and 50 mg alum/L was effective for turbidity reduction in most waters (Noblet 2012). Figure 4 shows the variation in percentage algal removal using an alum dose from 5 to 30 mg/L in pre-chlorinated waters. Applying 5 mg alum/L to the pre-chlorinated waters achieved an initial algal removal efficiency of 60% and 80% for the lowest and



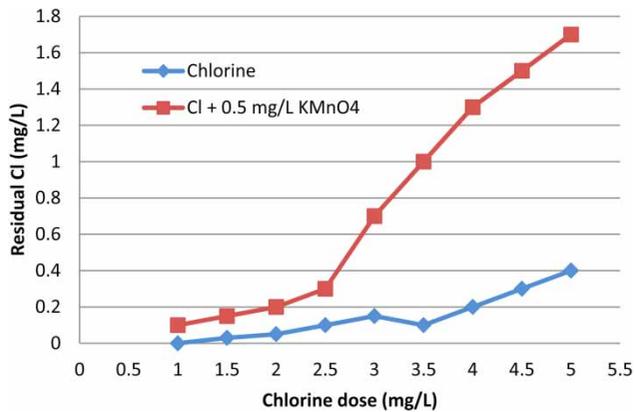
**Figure 4** | Mean algal removal (%) vs applied alum dose (mg/L). Mean seasonal algal counts: winter (4,000 units/mL) spring (3,000 units/mL), summer (3,200 units/mL) and fall (7,000 units/mL). Chlorine dose (mg/L): winter = 3.7, spring = 2.7, summer = 3.3 and fall = 5.5 (December 2011–November 2012).

highest algal counts, respectively. However, in order to achieve the 1.0 NTU WHO limit target, a higher alum dose of 30 mg/L and 25 mg/L was applied for 15 minutes and 30 minutes contact time, respectively. This achieved 86% and 89% reduction in final water turbidity for the respective contact times. In addition, these reductions were obtained for unbuffered waters and may benefit from the fact that alum addition to alkaline raw waters decreased the pH to the acidic range for an efficient coagulation and for better THM removal (Chen *et al.* 2009; Badawy *et al.* 2012; Noblet 2012). However, although alum is effective in removing turbidity, it is always applied in excess which may result in high residual Al: about 11% of the Al input remains in treated waters and may produce a residual concentration of 0.003–1.6 mg/L, which may exceed the acceptable limits (Wang *et al.* 2011).

#### Enhanced algal control and removal using combined chlorine/permanganate pre-oxidation

As indicated previously, while the major concern for the use of chlorine and its derivatives in water disinfection is the formation of chlorinated by-products, the application of  $\text{KMnO}_4$ , was reported to remove soluble manganese and iron which are frequently associated with the presence of cyanobacteria in raw water (Fan *et al.* 2013).  $\text{KMnO}_4$  may also serve as an alternative to pre-chlorination or other oxidants for the control of color, taste and odor, and algae and for the further control of biological growth in treatment plants. In addition, previous studies have indicated that combining chlorine with permanganate during the pre-oxidation stage has a synergistic effect upon water disinfection (Heng *et al.* 2008).

Previous studies have recommended the use 0.5–3 mg/L permanganate for enhanced algal removal by alum coagulation (Chen & Yeh 2005; Heng *et al.* 2008). Heng *et al.* (2008) indicated that a 0.5 mg/L permanganate dose in combination with chlorine achieved the best reduction efficiencies in turbidity and chemical oxygen demand<sub>Mn</sub> in raw surface waters. In the current study, the effect of adding an optimal constant dose of 0.5 mg/L  $\text{KMnO}_4$  upon the water chlorine demand was studied and the results are shown in Figure 5. It is evident from the results that the applied permanganate dose effectively reduced the chlorine



**Figure 5** | Effect of using  $\text{KMnO}_4$  (0.5 mg/L) on reduction in chlorine demand dose (mg/L) and the residual chlorine (mg/L) after application to raw surface water (initial mean algal count 7,000 units/mL during algal peak period).

demand and the resulting waters maintained an acceptable residual chlorine content required for bacterial growth control in the water supply delivery system. This may be explained by the synergistic inactivating effect and strong adsorptive capacity of the permanganate reduced product/intermediate ( $\text{MnO}_2$ ) (Shen *et al.* 2011). Also, the combination of permanganate/chlorine during the pre-oxidation stage may enhance the inactivation of algal cells and its removal through coagulation, an event that may control the production of THMs (USEPA 1999; Heng *et al.* 2008; Chen *et al.* 2009). Nonetheless, overdosing with permanganate must be avoided since a residual 0.05 mg/L  $\text{KMnO}_4$  or greater may result in a pink taint in the drinking water. Nonetheless, Fan *et al.* (2013) indicated that the application of a 1 mg/L dose  $\text{KMnO}_4$  may lead to a  $\text{KMnO}_4$  residual of 0.2 mg/L which becomes non detectable after 1–2 hours of treatment.

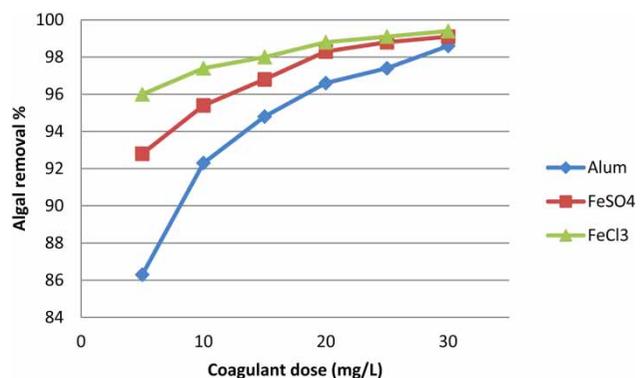
### The effect of the combined chlorine/permanganate pre-oxidation upon the use of Al and Fe coagulant salts

Aluminum sulfate, ferric chloride and ferric sulfate are the most common coagulants used in drinking water treatment (Lei *et al.* 2009; Matilainen *et al.* 2010; Sengul & Gormez 2013). The main purpose in using these coagulants is to reduce NOM which may interfere with the removal of other contaminants that cause membrane fouling, contribute to piping corrosion and act as a substrate for bacterial growth in the distribution system (Matilainen *et al.* 2010).

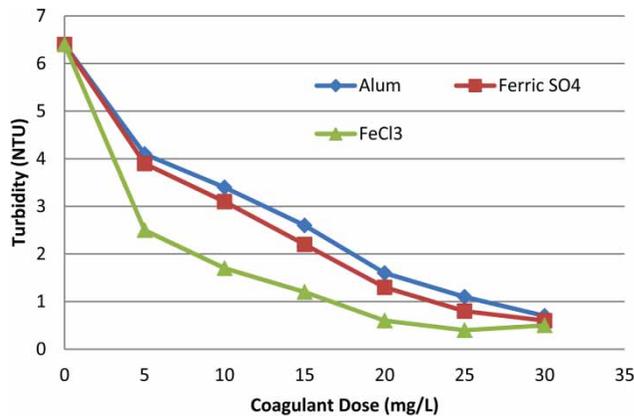
However, it is important to note that the action of aluminum and ferric salts depends upon their hydrolyzed form and their soluble complexes that possess high positive charge as well as upon an acidic pH range.

To confirm the effectiveness of these metal salts in coagulation using the combined permanganate/chlorine pre-oxidation, the residual turbidity and percentage algal removal of the raw and final treated waters were monitored after applying the coagulant doses (5–30 mg/L). Figure 6 shows that the application of these metal salts to Cl/ $\text{KMnO}_4$  pretreated raw waters achieved algal removals of over 90% at a dose as low as 10 mg/L. Figure 7 indicates that the maximum percentage algal removal and lowest residual turbidity was achieved using alum, ferric chloride and ferric sulfate doses ranging between 25 to 28 mg/L, 15 to 20 mg/L and 20 to 25 mg/L, respectively. Nonetheless, Al salts were reported to have a better capacity for turbidity removal and be more effective at lower doses than ferric salts, while ferric salts are more effective in NOM removal and are not as sensitive to temperature changes as alum (Matilainen *et al.* 2010).

Table 3 provides a summary of the finished water quality after the application of the combined pre-oxidation treatment for the raw water. Chen *et al.* (2009) reported that the presence of low calcium carbonate content (240 mg/L) in raw water was an important factor in the performance of the  $\text{KMnO}_4$ /alum system provided that the pH remained near alkaline. It was also reported that in the neutral pH range, surfaces of  $\text{MnO}_2$  and algae cells developed a negative charge and calcium ions in water served to bridge and



**Figure 6** | Effect of varying coagulant (alum, ferric chloride and ferric sulfate) doses upon the algal removal (%) in waters pretreated with a combined pre-oxidant dose of chlorine (2.7 mg/L)/permanganate (0.5 mg/L) during algal peak period.



**Figure 7** | Effect of varying coagulant (alum, ferric chloride and ferric sulfate) doses upon the final turbidity of water pretreated with a combined pre-oxidant dose of chlorine (2.7 mg/L)/permanganate (0.5 mg/L) during algal peak period.

**Table 3** | Effect of treatment by pre-oxidation and coagulants (alum, ferric sulfate and ferric chloride) at optimal dose on finished water quality parameters

Water quality parameters	Treatment			
	Pre-oxidation with 5.5 mg Cl <sub>2</sub> /L + 30 mg/L alum	28 mg/L alum	25 mg/L ferric sulfate	20 mg/L ferric chloride
pH	7.3	7.3	7.25	7.2
Turbidity (NTU)	0.33	0.3	0.3	0.25
Residual Al (mg/L)	0.23	0.17	0.05	0.05
Residual Mn (mg/L)	0.02	0.10	0.11	0.10
Residual Fe (mg/L)	0.07	0.00	0.14	0.13
Chloride (mg/L)	35	35	35	43
Sulfates (mg/L)	52	49.5	51.7	37
TOC (mg/L)	7.0	6.8	6.6	6.5
Algal count (unit/mL)	200	100	80	80

hold these negatively charged surfaces together into algal flocs (Chen *et al.* 2009). It can be seen from the data that ferric chloride and ferric sulfate provided slightly better finished water quality characteristics. However, the use of ferric chloride increased the final Cl as well as the use of alum and ferric sulfate increased the residual sulfates. Basiouny *et al.* (2008) studied the effect of KMnO<sub>4</sub> pre-oxidation alone upon the coagulation of raw Nile water. They found that an optimum KMnO<sub>4</sub> dose of 6 mg/L applied to Nile water resulted in a 35% turbidity removal. However,

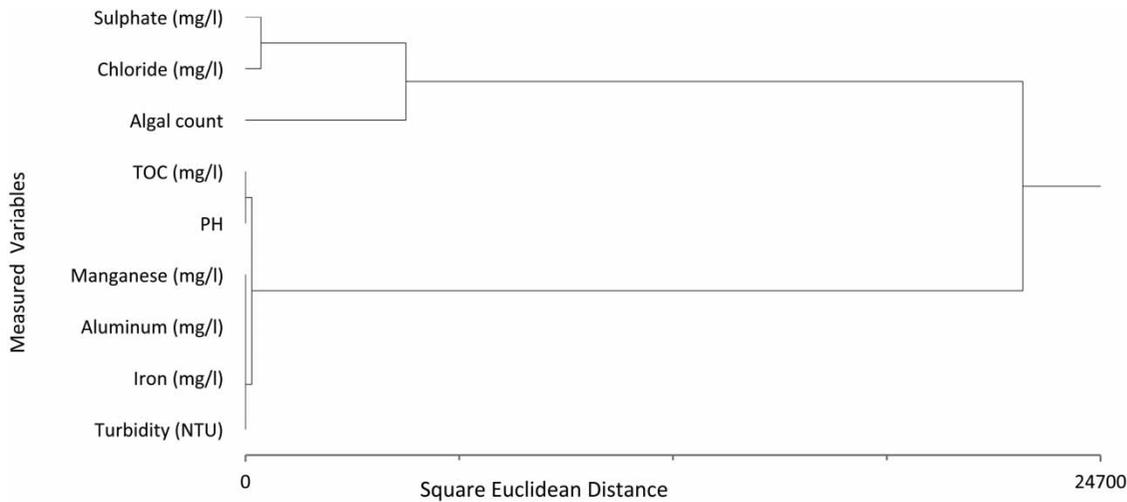
this required an alum dose of 40–60 mg/L and a ferric sulfate dose of 85–105 mg/L to achieve a turbidity reduction of 58% and 20%, respectively, with no pH adjustment.

Regarding the residual metal in final treated waters, accounts of the health effects and efficacy of using both Al and ferric salts in water treatment have been presented by Matilainen *et al.* (2010) and Simate *et al.* (2012). However, the use of alum as coagulant may lead to a high residual content as well, as it may form micro-flocs in the distribution networks which then provide nutrients for microbial growth (Lei *et al.* 2009; Matilainen *et al.* 2010; Sieliechi *et al.* 2010). On the other hand, it was reported that utilizing KMnO<sub>4</sub>/alum in combination did reduce the amount of residual alum in the finished waters (Tomperi *et al.* 2013). Ferric salts are also favored as they are more robust operationally and insensitive to the coagulation pH (Wang *et al.* 2013). However, ferric salts were reported to induce high alkalinity consumption plus the residual sulfates or chloride in finished water may increase its corrosivity (Matilainen *et al.* 2010). High residual iron content in drinking water may also be responsible for disinfectant neutralization, discoloration and a metallic taste in water (Sieliechi *et al.* 2010). As ferric chloride is a corrosive compound that could rapidly attack metals, treatment plants using it as coagulant may require major modifications in dosage equipments and system (storage tanks, pumps, piping, valves and accessories) (Niquette *et al.* 2004). In addition, the use of ferric sulfate in drinking water treatment may incur higher operational costs than using alum; however, it offers a higher satisfaction to the consumer (Niquette *et al.* 2004).

Hierarchical cluster analysis was performed on the final treated water main parameters and the results are shown in a dendrogram (Figure 8). From this figure, it can be seen that the reduction in algal count was dependent upon the sulfates and chloride content of the final treated waters. In addition, the reduction in TOC is strongly dependent upon the pH while the reduction in turbidity is more directly related to the hydrolyzed metal ions.

## CONCLUSION

The relationship between the Nile surface waters quality parameters and the occurrence of algae was investigated



**Figure 8** | Dendrogram depicting hierarchal clustering and distance between the average chemical and physical parameters of the final treated waters using Ward's methods for minimum variance.

using multivariate statistical analysis. The data revealed that Chlorophyta and diatoms contents were the major contributors towards the measured total algal count and that the Euglenophyta content was strongly dependent upon the total alkalinity ( $\text{CaCO}_3$ ) and the TDS of these waters. Cyanophyta also showed a strong dependency upon all chemical parameters measured as well as on Euglenophyta in these waters. As well, the effectivity of the current treatment procedure using pre-chlorination/alum on the clarification of the final treated water was investigated relative to the application of a combined  $\text{KMnO}_4$ /chlorine/coagulant stage. The results indicated that this application of  $\text{KMnO}_4$  was effective in reducing the chlorine demand while maintaining an acceptable residual content. The use of the combined chlorine/permanaganate pretreatment using alum, ferric chloride and ferric sulfate improved residual turbidity of the treated waters and enhanced percentage algal removal. Moreover, the finished water quality after the application of the combined pre-oxidation treatment for the raw water favored the application of ferric chloride and ferric sulfate. The study also revealed that the sulfates and chlorides ions contents were more responsible for the reduction in total algal count while the hydrolyzed metal ion was effective in the reduction of the final water turbidity. Economically, 1 kg of commercial permanganate costs 300 Egyptian pounds and 1 L of commercial  $\text{NaOCl}$  (14%) used at water treatment plants

costs 30 Egyptian pounds. However, without changing much of the current treatment system, the application of permanganate would be safer and reduce the health hazards associated with the chlorine addition.

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