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Surfactant Concentrations and Physicochemical Characteristics of Alexandria Coastal Water

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Abstract

The distribution of surfactant concentrations and the effect of physico-chemical characteristics (Temperature, pH, salinity, dissolved oxygen, oxidizable organic matter, alkalinity and calcium content) were studied. Water samples were investigated seasonally, during 2009 covering eight locations from El-Dekhila Harbor to Abu Qir Bay. The annual mean of water temperature and pH were 18.70- 27.11 °C and 7.82 – 8.48, respectively. The results revealed that the values of salinity, DO, OOM, alkalinity and Ca were given as annual mean in the ranges: (33.75- 39.10; 37.38 ± 1.53 ‰), (1.32-5.18 ; 3.73 ± 0.99 ml/l), (0.32- 10.56; 5.00 ± 3.12 mg/l), (2.52- 3.21; 2.74± 0.22 meq/l), and (382- 443; 416.65 ± 18.54 mg/l), respectively. The results of linear alkyl benzene sulfonate "LAS" concentrations revealed a high range of variation and regional irregularities (0.088 – 0.333; annual mean 0.188± 0.095 mg LAS/l). The study indicated that the mean concentrations of surfactant examined here are higher than those reported for unpolluted Seawater (0.01 LAS mg/l) and typical of open ocean water.

Key words: Alexandria, coastal area, physical parameter, surfactant

1. Introduction

Surfactants are a diverse group of chemicals that are designed to have cleaning or solubilization properties. They generally consist of a polar head group (either charged or uncharged), which are well solvated in water, and a non polar hydrocarbon tail, that not easily dissolved in water. Hence, surfactants combine hydrophobic and hydrophilic properties in one molecule. The world production of synthetic surfactants amounts to 7.2 million tons annually (Di Corica, 1998). Linear alkyl benzene sulfonates (LAS) are the most popularly used anionic surfactants, and extensively used for over 30 years with an estimated global consumption of 2.8 million tons in 1998 (Verge *et.al*., 2000). LAS is used in nearly all kinds of detergents and cleaning preparations including laundry powders, laundry liquids, manual dishwashing liquids, detergent tablets, soap bars, and household cleaners**.** In order to assess their environmental risks, we need to understand distribution, behavior, fate and biological effects of these surfactants in the environment. Theoretically, the surfactants can be degraded and removed by chemical, physical and biological processes in wastewater treatment plants (Swisher, 1987). However less than 5% of all municipal wastewater is treated by wastewater treatment plants in Egypt. Large quantities of surfactant residues in wastewater are discharged into the lakes, rivers and seawaters. Pollution problems of the hot spots (El-Dikheila harbor, El-Mex Bay, Western harbor, Eastern harbor and Abu Qir Bay) in Alexandria are well known, but no enough studies have reported the distribution and concentrations of surfactant residues and their degradation products in the aquatic environment.

The biodegradation of LAS is affected by a numerous factors amongst which are the concentration of dissolved oxygen (Krueger *et.al*., 1998), complexion with cationic surfactants (Utsunomiya *et.al*., 1998) and the formation of insoluble calcium and magnesium salts (DeWolf and Feijtel, 1998), the presence of other organic contaminants (Abd-Allah and Srorr, 1998), temperature (George, 2002) and the effect of LAS on the pH during aerobic degradation (Garcia *et.al*., 1996). In sewage-contaminated groundwater the rates of LAS biodegradation increase with dissolved oxygen concentration and the longer alkyl chain homologues (C_{12} and C_{13}) are preferentially biodegraded. Many investigators had studied the hydrographical, trace metals and chemical characteristics of Alexandria coastal seawater and sediments as well as phytoplankton standing crop; (Abd-Allah, 1995; Mahmoud, 1989, 1991; El-Sherif and Mahmoud, 1991; El-Deeb and Emara, 2005; Okbah, 2006). The aim of the present work is to study the regional and seasonal variations of commercial anionic surfactants (LAS) in seawater as well as physicochemical characteristics of Alexandria coastal area from El-Dikheila harbor to Abu Qir Bay.

2. Material and Methods

2.1. Study Area

The investigated area lies between latitude 31º 8' & 31º 17' N and longitude 29º 47' & 30º 4' E (Figure 1) which shows also the location of sampling stations. It comprises most of the coastal area of Alexandria, extending from El-Dikheila Harbor at the southwest to Abu Qir bay at the northeast with a shoreline of about 28 kilometers long. The area includes three harbors; the two old harbors (the Western and Eastern Harbors) and the newly constructed El-Dikheila Harbor. Also, the study area includes El-Mex and Abu Qir Bays, El-Mex bay lies at the western region of Alexandria City which represents semi-closed shallow basin with an average depth of about 9.0 m bordered from its northeastern side by the Western Harbor outlet and by El-Dikheila Harbor at the northwest. Its southern side is El-Mex area which receives about 2.4 x 10^9 m^3 per year of drainage, sewage and industrial wastewaters from Lake Mariut through El-Mex Pumping Station (El-Deeb and Emara, 2005). Abu Qir Bay is a shallow basin lying 35 km east of Alexandria City; it receives various types of wastewaters from El-Tabia Pumping station, Boughaz El-Maadia, and Rosetta at the mouth of the Nile River. It has an area of 500 km^2 with an average depth of 12m and a shore line of 50 km long. Approximately 3.3 million m^3 d^{-1} of brackish water is introduced into the Bay from Lake Edku through Boughaz El-Maadia (Okbah, 2006).

2.2. Sampling and Analysis

Eight surface seawater samples were collected seasonally (Figure 1) from Alexandria coastal area using Nisken bottle during the period from March 2009 to January 2010. Hydrographic and some chemical parameters (temperature, pH and dissolved oxygen) were measured in-situ. Water temperature and pH were measured directly in the field using a standard Schmidt thermometer and a portable digital pH-meter (model 210), respectively. Salinity was determined using a Beckman Induction Salinometer (model RS-7C). The classical Winkler method, modified by Grasshoff (1983) used for determination of dissolved oxygen (DO). Determination of total alkalinity was carried out according to Riley and Skirrow (1961). Calcium (Ca) concentration was determined by the EDTA titration. The method used for oxidizable organic matter determination was described by FAO (1976)

2.3. Analysis of LAS in seawater

Rapid determination of anionic surfactants (LAS) by improved spectrophotometer method using methylene blue (MB), as cationic dye, was investigated by Koga *et al*. (1999). The method based on the reaction between linear alkyl benzene sulfonate (LAS), as anionic surfactants, and methylene blue (MB), as cationic dye, to form associated ion-pair (LAS-MB ion pair) in water with 1:1 molar ratio which can be easily extracted to the organic phase (chloroform, $CHCl_3$).

3. Results and discussion

3.1. Physico-chemical Characteristics of Coastal Seawater

3.1.1. Water Temperature

The hydrographic conditions and surfactant concentration varied widely during the study period. The water temperature did not deviate from the seasonal fluctuations normal on Egypt's Mediterranean coast, the maximum temperature attained in August reaching 27.11 ºC while the minimum temperature was about 18.10 ºC in January. These values are similar to that reported by Dorgham *et al*. (2004). Spatial variations in the surface water temperature were recorded. The largest difference of water temperature for the study area was reported in spring and autumn (2.13 and 2.45 ºC, respectively.) and the smallest was in summer and winter (0.94 and 1.60 ºC, respectively.).

3.1.2. pH

The values of pH of the investigated area lie on the alkaline side, the pH values ranged between 7.82 (St.I; Dekhila Harbor) and 8.48 (St.II; Western Harbor) (Table 1 and Figure 2). The pH values may be controlled by the density of phytoplankton standing crop and the water quality inflow to the study area.

3.1.3. Water Salinity (PSU)

Salinity is an important factor which reflects the changes caused by the mixing of fresh water, drainage water and seawater. The regional variation value of water salinity was recorded in Table (1) and Figure (3). The lowest value (33.75 PSU) was recorded at (St.VII) in Abu Qir Bay, which is affected by the wastes discharged, while the highest one (39.10 PSU) was recorded at (St.VI) in NIOF, which is distance away from the impact of the discharged wastes (Figure 2). The present data of water salinity showed slightly variation in summer season (around 38 PSU) and autumn season (around 37 PSU). On the other hand, the values of water salinity revealed wide variations recorded in spring (ranged; 33.75 - 38.01 PSU) and winter season (ranged; 36.13 – 39.10 PSU).

3.1.4. Dissolved oxygen (DO)

The regional and seasonal variations of DO are recorded in Table (1) and Figure (2). Wide variation was obtained between minimum and maximum values Figure (2). The absolute value of DO (Table 1)

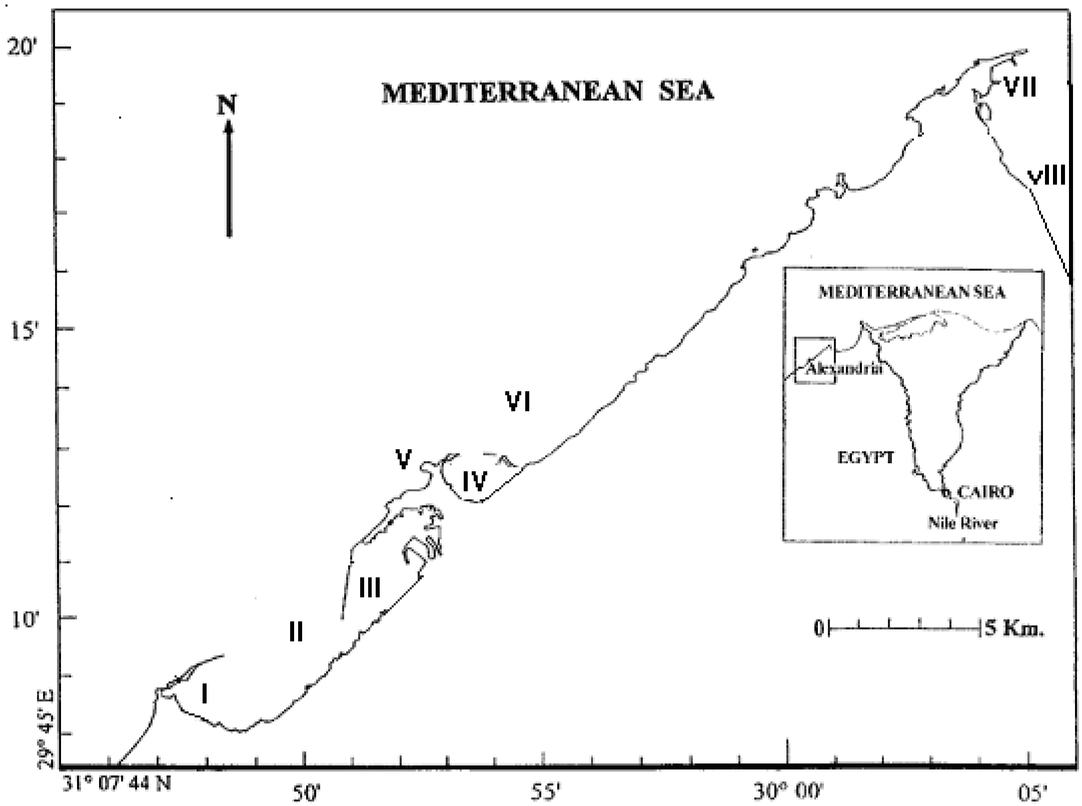

Figure 1: Study Areas

(I: Dekhaila Harbor, II: Western Harbor, III: El – Mex, IV: Eastern Harbor (II), V: Eastern Harbor (I), VI: NIOF, VII: Abu Qir, VIII: El – Maadia)

Table 1: Range and Average Values of Physico-chemical Properties and LAS concentration Along Alexandria coastal zone

Season	Spring		Summer		Autumn		Winter	
Parameter	Range	AVE ± SD	Range	AVE ± SD	Range	AVE ± SD	Rang	AVE ± SD
LAS (mg/l)	0.09 - 0.19	0.14 ± 0.03	0.1 - 0.33	0.21 ± 0.08	0.16 - 0.23	0.21 ± 0.07	0.08 - 0.33	0.2 ± 0.07
pH	7.91 - 8.2		8.21 - 8.48		7.98 - 8.2		7.82 - 8.2	
Temp. °C	18.7 - 20.83	19.46 ± 0.82	26.17 - 27.11	26.83 ± 5.96	19.08 - 21.53	20.02 ± 5.58	18.1 - 19.7	18.71 ± 5.37
S (PSU)	33.75 - 38.01	36.55 ± 1.73	38 - 38.2	38.13 ± 7.55	36.7 - 37.3	37.13 ± 7.8	36.13 - 39.1	37.85 ±7.95
DO (ml/l)	3.82-5.19	4.61 ± 0.51	1.24 – 3.14	2.29 ± 1.42	3.00– 4.71	3.86 ± 1.28	4.07 – 4.63	4.28± 1.21
OOM (mgO_2/l)	0.32 - 6.24	2.66 ± 2.28	5.56 - 10.32	8.1 ± 3.39	2.56 - 7.68	5.25 ± 2.94	4.08 - 8.56	6.23 ±2.73
Alkalinity (meq/l)	2.63 - 2.98	2.8 ± 0.12	2.6 - 3.21	2.8 ± 0.59	2.54 - 2.81	2.66 ± 0.59	2.52 - 3.01	2.71 ± 0.6
Ca (mg/l)	382 - 443	413.8 ± 23.8	390 - 435	415.8 ± 84.1	397 - 442	422.4 ± 87.9	389 - 439	414.1 ± 88.9

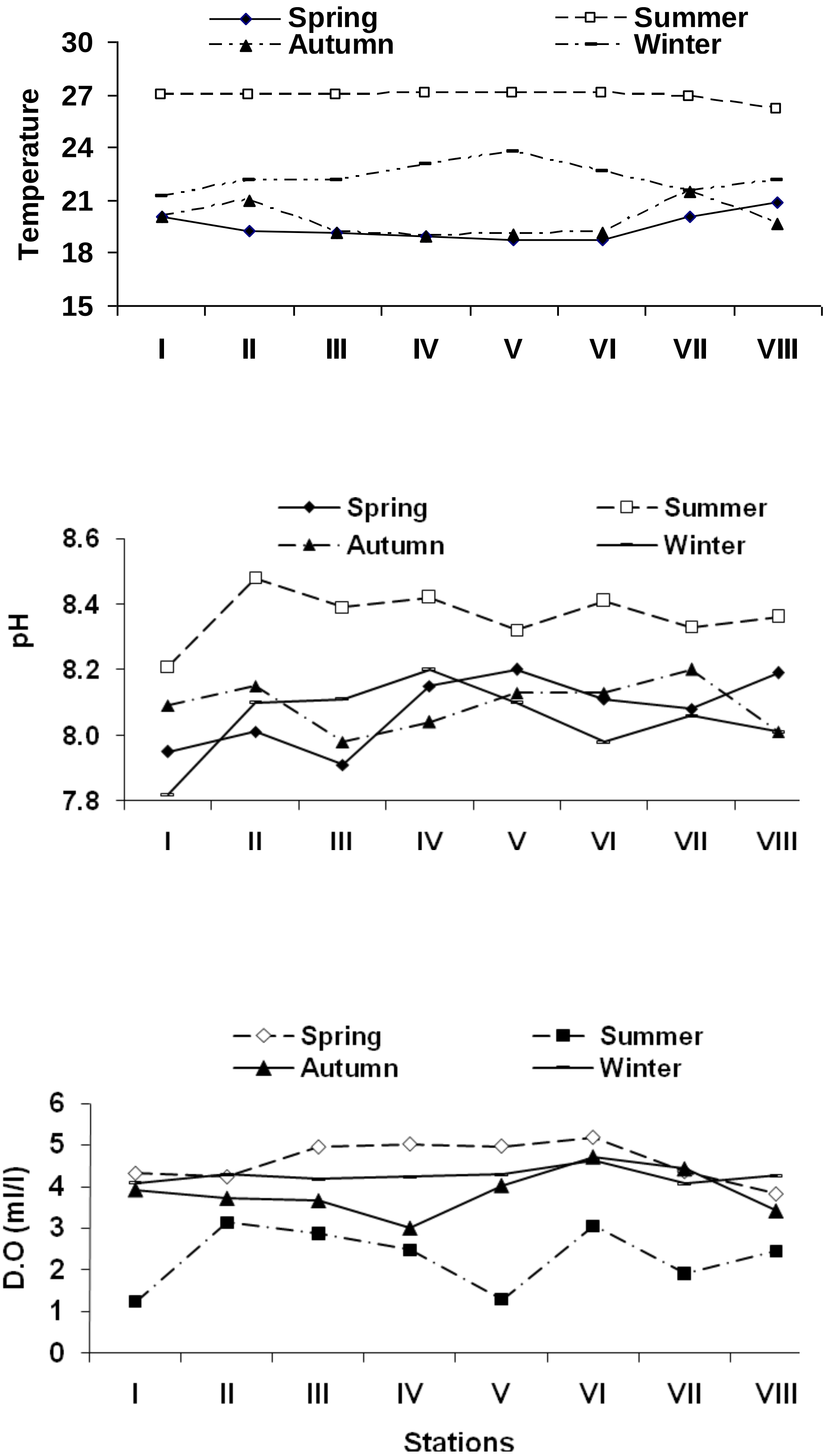

Figure 2: Regional and Seasonal Variations of Temperature, pH-Values and Dissolved Oxygen (D.O.) along Alexandria Coastal Area.

fluctuated between 1.239 ml l^{-1} (St.I; Dekhila Harbor) in summer 2009 and 5.187 ml l^{-1} (St.VI; NIOF) in spring 2009. In the study area, the DO content of seawater during summer season was relatively low (1.239 – 3.143 ml l^{-1}) compared with spring and winter seasons (3.892 – 5.187 ml l^{-1}). The low values of DO may be attributed to respiration of marine organisms and biochemical transformations of organic matter and mainly due to high temperature in summer. The most important factors controlling the DO budget were the quantity and quality of the discharged sewage wastes, the exchange of water with the adjoining open Mediterranean waters and the high rate of photosynthetic activity of phytoplankton production that produced excess of oxygen. These conditions were clearly demonstrated in the surface water of the study area during the year, when the lowest surface dissolved oxygen content in summer and autumn seasons was accompanied with the highest surface oxidizable organic matter in the same seasons

3.1.5. Ca^{+2} *and Total alkalinity*

In the aquatic environment, the Ca^{+2} is a major cation which is the necessary element for the growth of phytoplankton and for aquatic animals (Conley, 2000). The regional and seasonal distributions of Ca^{+2} and total alkalinity are presented in Figures (3&4) and Table (1). The seasonal average values of Ca^{+2} fluctuated in the range of 218±135 mgl^{-1} in winter and 251±162 mgl^{-1} in spring season. Negative correlation was observed between Ca^{+2} and LAS concentrations (r= -0.451, n=32, P<0.05) due to precipitation. Calcium ion concentrations play an important role in the solubilization or precipitation of LAS from seawater column to reflect the bioavailability fraction of LAS for aquatic organisms and consequently the toxicity reduction (Verge *et al*., 2000)

Total alkalinity is simply expressed as the sum of equivalents of HCO_3^-, CO_3^- and $Ba(OH)_4^-$ ions. In general, carbonate and bicarbonate ions are the main components contributing to the alkalinity in most natural waters. The values of total alkalinity fluctuated between 2.52 $meql^{-1}$ at (station VI; NIOF) and 3.21 $meql^{-1}$ at (station V; Eastern Harbor). The seasonal average values of total alkalinity varied between 2.66 ± 0.59 $meql^{-1}$ in autumn and 2.80 ± 0.12 $meql^{-1}$ in spring and summer seasons. The high alkalinity of the surface water might partially result from the large amount of freshwater discharged to the study area. This evidence can be supported by the negative correlation obtained between total alkalinity and salinity in the surface waters of the study area (r=-0.321, n=32, P<0.05). The effect of total alkalinity on LAS concentrations was indicated by the inverse correlations between the two variables (r=-0.311, n=32, P<0.05).

3.1.6. Oxidizable organic matter (OOM)

Measurement of the oxygen equivalent to the amount of material oxidized by a strong oxidizing agent, such as potassium permanganate ($KMnO_4$) gives a convenient characterization of the water quality. High values of oxidizable organic matter (OOM) indicate water pollution, which is linked to sewage effluents discharged to the area. In the present study, the concentrations of the oxidizable organic matter showed seasonal and regional variations. The values ranged between a minimum of 0.32 at (St.V Eastern Harbor) in spring and a maximum of 10.32 mg O_2/l at (St. VIII El-Maadia) in summer (Table1). The seasonal average concentration fluctuated from 2.66±2.28 mg O_2/l in spring to 8.10 ±3.39 mg O_2/l in summer season.

The concentrations of organic matter in the surface waters of the study area were noticeably high in the warm seasons and low in the cold ones Figure (3) due to increases of human population and activities during summer seasons. The effect of temperature, as a dominant factor accelerating the rate of organic matter oxidation was indicated from the direct associations between the $KMnO_4$ consumption values and water temperatures (r=0.35 n=32 <0.05) in the surface water of the study region. The previous studies showed the effect of organic matter (humic substances) on LAS bioavailability. The toxicity of LAS with *Pimephales Promelas* was studied by McAvoy *et al.* (1996); their results (2.5 – 55 mg/l) revealed that LC50 at 96 h increased from 1 to 2 mg/l by adding different quantities of humic acid.

3.2. Distribution of Surfactant along the study area

Theoretically, the surfactants can be degraded and removed by chemical, physical or biological processes in wastewater treatment plants (Ding *et al.*, 1999) ·However, lesser than 5% of all municipal wastewater is treated by wastewater treatment plants in Alexandria. Large quantities of surfactant residues in wastewaters are discharged into the seawater directly (Nazih, 2006).

The concentration of anionic surfactant LAS in the coastal zone water of Alexandria City are given in (Table 1). The regional and seasonal variations of anionic surfactant LAS at the surface water Figure(4) , depicted that stations III, IV, V and VIII had the maximum LAS concentration values 0.333 and 0.305 mg LAS l^{-1} for stations III and VIII, respectively in summer season and 0.327 and 0.293 mg LAS l^{-1} at station IVand V, respectively in winter season. The relative high levels are attributed to wastewater discharge into the coastal area. The amount of drainage water of Alexandria City discharged into the sea fluctuates between 4.8 and 7.1 million cubic meters per day with an annual average of about 2,420 million cubic meters (El-Sherif and Mahmoud, 1991), so that the concentrations decreased significantly with the distance from the point of discharge. On the other hand, the lowest LAS concentrations were 0.092, 0.082 and 0.087 mg LAS l^{-1}, which are recorded in

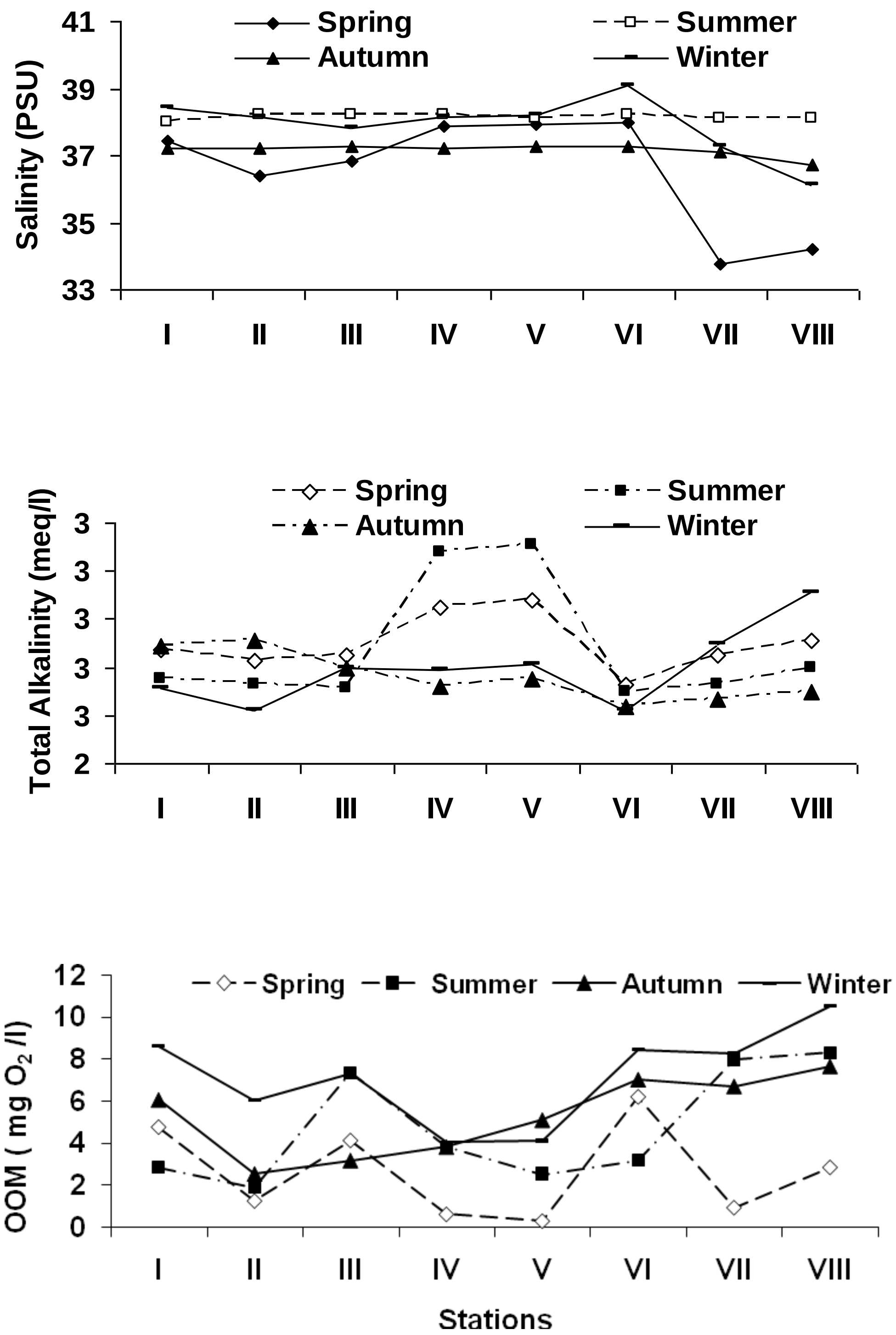

Figure 3: Regional and Seasonal Variations of Salinity, Total Alkalinity and Oxidizable Organic Matter (OOM) along Alexandria Coastal Area.

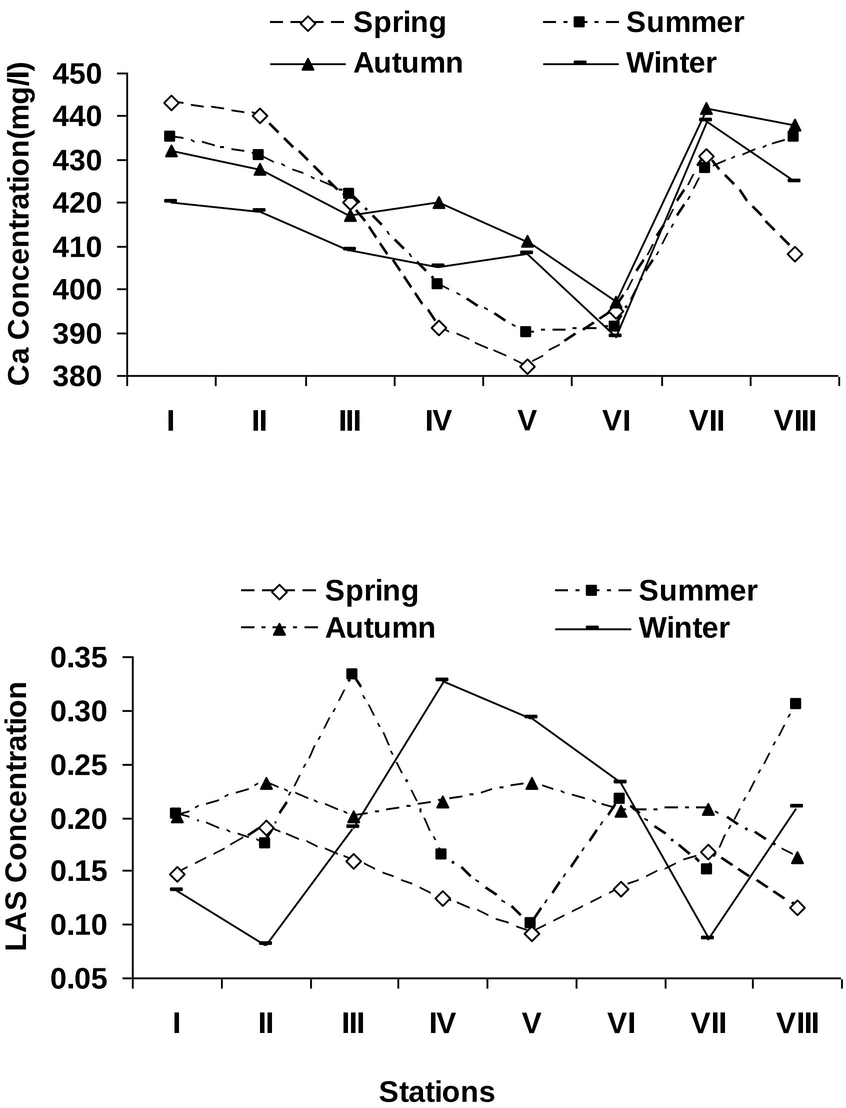

Figure 4: Regional and Seasonal Variations of Calcium Concentration (mg/l) and Surfactant content (mgLAS/l) along Alexandria Coastal Area.

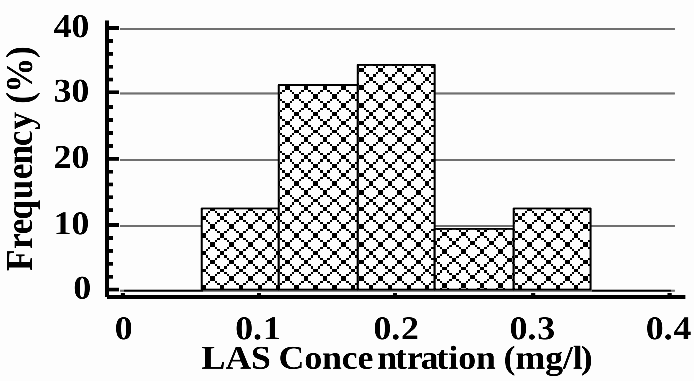

Figure 5: Percent Frequency Distribution of LAS in Alexandria Coastal Area

spring (stationV) and winter (stations, I and VII),respectively. In general, the presence of surfactants in all cases was related to sewage disposal in the system, and the greatest concentrations of these compounds were found near the source of discharge.

The biodegradation processes were shown to be seasonally dependent (George, 2002). The seasonal average of linear alkyl benzene sulphonates, LAS, in Alexandria coastal zone area during period of study revealed irregular variations Figure (4). It is observed that the summer had relatively high concentration, its content ranged from 0.10 to 0.33 (average; 0.21±0.08) mg LAS/l. This value is similar to that reported in autumn season (0.16 and 0.23) (average; 0.21±0.07) mg LAS/l. At the temperature representative of summer the transformation of LAS is faster than winter temperature conditions (George, 2002). The highest concentration in summer is attributed to increase in population density and human activities in this area which lead to increase in domestic loading during this season. In winter and spring, LAS concentrations showed the lowest Figure (4), with values ranging between 0.08 and 0.33 (average; 0.20 ±0.07) mg LAS/l and from 0.09 to 0.18 (average; 0.14 ±0.03) mg LAS/l in spring. There is no clear relationship between LAS concentrations and water temperature, for the first and second season respectively probably resulting from continuous input of wastewater and surfactants to this area. This seasonal variation is related to the amount of urban drainage, industrial waste discharged that contains detergents.

The relationship between LAS concentrations and salinity as a conservative parameter, which show the extent of mixing of seawater with sewage discharge, was calculated. There is a significantly high negative correlation (-0.82) showing that with increasing S‰ the value of LAS decrease.

According to Holt *et al.* (1989), the decrease in LAS concentration with the distance from the source of contamination was faster than that predicted based only dilution. It is anticipated that removal mechanisms from the seawater column include biodegradation (Rego, 1998), sorption for suspended solids (Westall *et al.*, 1999), and precipitation with divalent cations (Xie *et al.*, 1997). Since sorption of LAS on sediment is promoted when Ca^{2+} concentration increases (Westall *et al.*, 1999). A tentative comparison between LAS concentrations and calcium ion concentrations (water hardness) during investigation period of study were calculated. Water hardness also can be used as indicator variable showing the mixing between the brackish water and seawater in the study area. There were a significantly high negative correlation between LAS contents and calcium ions concentrations ($r=-0.43, n=32, p<0.05$) indicating thus strong precipitation of LAS as $Ca(LAS)_2$ when Ca^{2+} concentration increases and seems to promote cooperative sorption at high surfactant and calcium ion concentrations (García *et al.*, 2002) and the affinity for the anionic surfactant after an initial adsorption. Calcium ions could adsorb directly to the sludge particle, yielding positively charged sites onto which negatively charged LAS homologues can adsorb. Verge *et al.* (2000) introduced the effect of LAS precipitation, due to water hardness, on bioavailability and consequently on toxicity. The biological test used was the acute toxicity to *Daphnia*. Calcium precipitation boundary diagrams are very useful tools to evaluate anionic surfactants-calcium ion interactions

It is observed that the detergents content of the Red Sea coastal water in front of Hurgada was 0.1-0.26 mg LAS l^{-1} (Mahmoud, 1998),it is remarkably lesser than its content in the Eastern Harbor (0.05-3.06 mg. LAS l^{-1}) (Mahmoud ,1989). The present data are lesser than that recorded by El-Sherif and Mahmoud, (1991) and Mahmoud, (1991) in El-Mex Bay (0.08-1.70 mg LAS l^{-1}) and Abu-Qir Bay; 0.0-4.71 mg LAS l^{-1}), respectively. These results are similar to the results of the brackish water of Lake Borollos in Nile Delta (.0-0.17 mg LAS l^{-1}) (Beltagy and Mahmoud, (1988) but higher than those recorded at El-Agami; 0.02-0.08 mg LAS l^{-1}(Mourad. and Abd-Allah, 1995). Lake Maryout represents the highest contaminated area in Alexandria City (3.6-18.6 mg LAS l^{-1}) Mourad and Abd-Allah, 1995).The high level is attributed to the huge amount of industrial and sewage discharge to these areas. El-Sherif and Mahmud (1991) reported that, the anionic detergents concentration had negative effect on the standing crop of phytoplankton in El-Mex Bay. The seasonal variations of El-Mex Bay surface water showed an outstanding peak of phytoplankton during spring and it consisted mostly of green algae, which was accompanied by low concentration of detergents. Lower counts of phytoplankton were recorded in the other seasons. The sharp drop of phytoplankton counts recorded in summer was accompanied by rapid increase of detergent concentration.

Figure (5) shows the distribution of frequency percent of LAS concentration in Alexandria coastal area. It is cleared that the most abundant values ranged between 0.1 and 0.2 mg LAS/l which represent about 64 % of the total samples. The high values of LAS content ranged between 0.2 and 0.3 mg LAS/l representing about 24 % of the total samples, while the low values lesser than 0.1 mg LAS/l constitutes about 12 % of the total samples. The frequency distribution of the concentration of methylene blue active substances in the Eastern Harbor area was ranged between 0.0 and 0.8 mg LAS/l represented about 68% of the total samples (Mahmoud, 1989).In the investigated area, the levels of surfactant are significantly higher at the Eastern Harbor and Abu Qir Bay by approximately 4 and 6 fold compared with data recorded by Mahmoud (1989). The acute toxicity concentration of the most commonly used of surfactant recorded by Albaster (1978) was ranged from 3 to 7 mg LAS/l. Comparing with the results recorded by Kozarac *et al*. (1977) in unpolluted sea water (lesser than 0.01 mg

LAS/l),depicted that the concentrations in offshore marine waters are less than 0.001-0.002 mg/l (WHO, 1996). From these data it is evident that the level of surfactant in the study area is high.

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دراسة الخصائص الفزيقوكيميائيه و تركيزات سلفونات الإلكيل بنزين الخطيه فى المياة الساحلية بالاسكندريه-مصر

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تم دراسة توزيع تركيزات سلفونات الإلكيل بنزين الخطيه فى المياة الساحلية بالاسكندريه (المنظفات) وتأثير الخصائص الفيزيائية والكيميائية (درجة الحرارة ، درجة الحموضة والملوحة ، الأكسجين الذائب ، المواد العضوية القابلة للأكسدة ، والقلوية والكالسيوم). وقد تم جمع عينات المياه موسميا خلال عام 2009 التي تغطي ثمانية مواقع من ميناء الدخيلة الى خليج أبو قير. وكان المعدل السنوي لحرارة المياه ودرجة الحموضة 18.70 -27.11 درجة مئوية و 7.82 حتي 8.48 على التوالي. وكانت القيم المتوسطة وكذلك مدى هذه التركيزات لكل من قيم الملوحة والاكسجين الذائب والمواد العضوية والقلوية والكالسيوم الذائب كالتالى : (39.10-33.75 بمتوسط 1.53± 37.38 ‰) ، (5.18-1.32 بمتوسط 3.73 ± 0.99 مل / لتر) و (0.32- 10.56 بمتوسط 5.00 ± 3.12 مجم / لتر) و (3.21-2.52 بمتوسط 2.74 ± 0.22 مل مكافئ / لتر) و (382 - 443 بمتوسط 416.65 ± 18،54 مللجم / لتر) على التوالي. وقد اظهرت النتائج لتركيزات سلفونات الإلكيل بنزين الخطيه تركيزات عالية تغيرات بطول منطقة الدراسة و تراوحت مابين (0.088-0.333 بمتوسط سنوى 0.188±0.095 مللجم لاس/لتر) وأشارت الدراسة إلى أن متوسط تركيزات سلفونات الإلكيل بنزين الخطيه التى درست هنا أعلى من تلك التي ذكرت فى مياه البحر الغير ملوثة (0.01 لاس مللجم / لتر).