ISSN 1110-0354
Bull. Nat. Inst. of Oceanogr. & Fish., A.R.E., Vol. (27) 2001: 45-63

TOTAL ALKALINITY, CARBONATE AND SPECIATION OF BICARBONATE IN FRONT OF ALEXANDRIA, EGYPT

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Key words: Alkalinity, bicarbonate, carbonate, lake water, seawater.

ABSTRACT

During the period from February to August 2000, surface water samples were collected from five different locations, four from the Mediterranean inshore waters along the Alexandria coast (Eastern Harbour, Kayet bey, El-Mex bay and El-kilo 21), while the last is from Lake Edku. These regions are subjected to land-runoff. Samples were analyzed for total alkalinity, carbonate and bicarbonate. Calculation of specific alkalinity was done. The concentrations of bicarbonate varied between 184.42 and 428.42 mg l for Lake Edku and 111.30 to 326.28 mg l for the inshore waters. The results were interpreted in terms of factors affecting the sites. For both Lake Edku and the inshore waters, the maximum average ratios of specific alkalinity (12.737±0.580 and 0.615±0.120 respectively) were recorded at regions affected by the drainage water.

Computer equilibrium modelling was also done, and the calculated bicarbonate species were discussed. The modelling studies show that the free ion HCO_3^- *and the ion pairs* $NaHCO_3^0$ *and* $MgHCO_3^-$ *are the main forming bicarbonate species. In all the studied waters, the free ion* HCO_3^- *is the dominant species.*

INTRODUCTION

Total alkalinity of a natural water sample can be regarded as a measure of the proton deficit of the solution relative to an arbitrarily defined zero level of protons (Dickson, 1981). It is affected by several processes which are photosynthesis and respiration, nitrification, denitrification, sulfide oxidation, sulfate reduction, and $CaCO_3$ dissolution (Stumm and Morgan (1996)). Measurement of water alkalinity deserves to be studied extensively as; alkalinity specific alkalinity may be a potentially useful complementary tracer for studying mixing processes in coastal areas where seawater mixes with outflows from multiple rivers (Wong, 1988). Alkalinity, is very closely associated with the forms of carbon dioxide system in aquatic environments (aquated CO_2, carbonic acid (H_2CO_3), bicarbonate ions (HCO_3^-) and carbonate ions(CO_3^{2-})).

In Egypt, various studies have focused on the distribution of alkalinity in marine waters (Tayel *et al* 1996, Tayel and Shriadah 1992, Shriadah and Emara 1992, and Dowidar *et al* 1987) and in lacustrine environments (Tayel 1992, Abdel-Moneim *et al* 1987, and Mahlis *et al* 1970). Forms of alkalinity (carbonate and bicarbonate) in different aquatic environments have received widespread attention by several researchers (Youssef 1999, Abdel Moati 1985 and 1981, and Levy 1974).

The main purpose of the present study is to discuss total alkalinity, specific alkalinity, carbonate and bicarbonate levels in some Mediterranean coastal waters subjected to land-runoff (inshore waters along the Alexandria coast beside the coastal Lake Edku). The second aim is to describe the speciation of bicarbonate in these environments.

MATERIALS AND METHODS

The Eastern Harbour (E.H.) is a semi-enclosed area with its mouth protected from the sea by an artificial break water barrier leaving two openings to the sea, namely El-Boughaz and El-Silsila. Its surface area of about 2.8 km^2 and its average depth is about 5 m, E.H. has been affected by sewage waste disposal through many outfalls. Kayet Bey receives daily ~ 300,000 m^3sewage water through its main disposal site (Map 1). El-Mex Bay (Map 2) extends for about 15 km between El-Agamy headland in the west and the Western Harbour in the east, with a mean depth of 10 m. It receives huge volumes of drainage water via El-Umum Drain (about 10 x $10^6 m^3 day^{-1}$, personal communication). The region of El-kilo 21 west of Alexandria (Map 3) is receiving drainage water from Noubaria Canal.

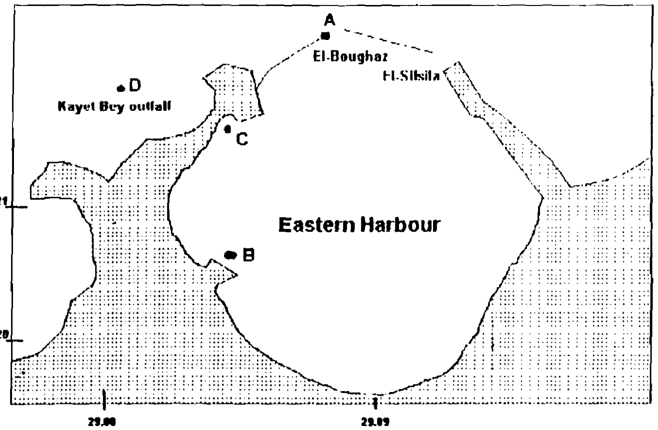
Lake Edku (Map 4) is one of the four Northern Nile Delta Lakes. It lies on the Mediterranean coast, west of the Rosetta branch of the Nile. The Lake covers a total area of about 126 km^2; most of it, particularly its eastern side, is densely vegetated, leaving a water-free area of about 79 km^2. The depth varies between 0.4 and 1.5 m, with an average of about 1 m.

In the course of the investigation from February to August 2000, February, May, July and August surface water samples, representing four locations, were collected from the coastal waters of the Mediterranean Sea, Alexandria. Figures 1-3 show the locations under study which include the Eastern Harbour (stations A-C), Kayet Bey (station D), El-Mex Bay (station E) and at El-kilo 21 (stations G, H) as well as the mixing sites between the Drains (El-Umum Drain (station F) and Noubaria Canal (station I)) and the sea water. Station K represents water of El-Umum Drain and station J represents that of El-Noubaria to some extent affected by seawater. Samples were also collected from 10 stations, representing the whole area of Lake Edku, as well as from the connected Drains El-Khairy and Barsiek (Map 4).

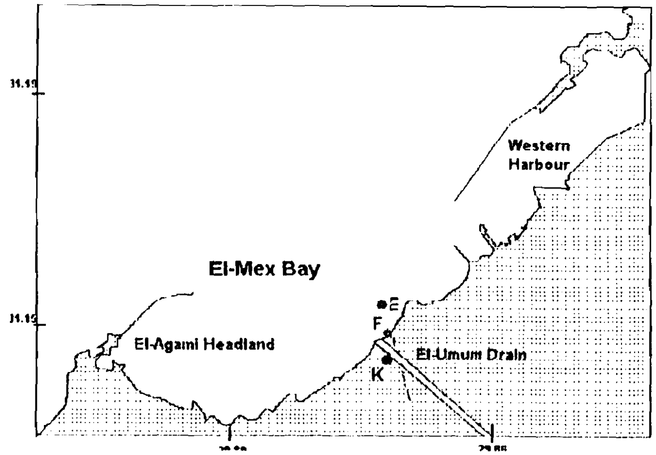
Determination of total and carbonate alkalinity were carried out according to the method described in APHA 1985. Bicarbonate alkalinity = total alkalinity – carbonate alkalinity. Specific alkalinity was calculated according to Strickland and Parsons (1972). Chloride values were taken from Youssef. and Lees-Gayed and Abbas and Youssef for seawater and Lake Edku respectively (in press).

Speciation of bicarbonate was done using the program WATEQP version 2.0 (Appelo and Postma 1994). This program implements an ion association equilibrium model, using the equilibrium constants of specified association reactions to calculate the activities and activity coefficients of the different species in water samples whose analyses are used as input. The equilibrium constants were obtained almost entirely from the NIST Critical Stability Constants Database version 4.0 (Martell *et al* 1997). The values of temperature and major cations (sodium, potassium, calcium and magnesium) were taken from Youssef. and Lees-Gayed (in press). Activity coefficients were calculated using the Davies equation:

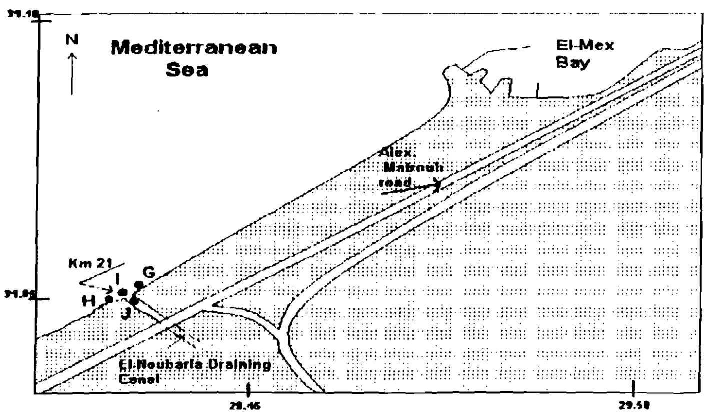
$$\log \gamma_i = -A z_i^2 \left(\frac{\sqrt{I}}{1+\sqrt{I}} - 0.3\, I \right)$$



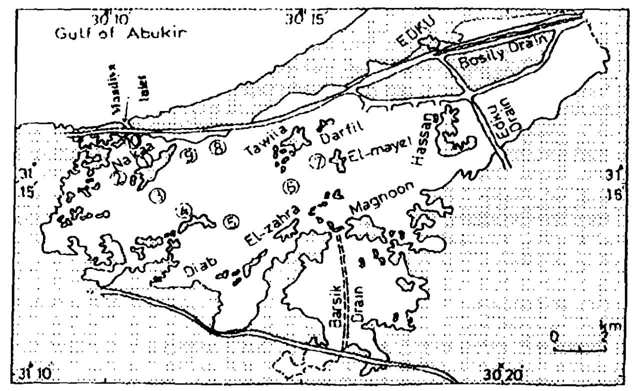
Map 1: Eastern Harbour and Kayet Bey



Map 2: El-Mex Bay



Map 3: Region of El-kilo 21



Map 4: Lake Edku

where γ_i is the activity coefficient of an ion i of charge z_i in a solution of ionic strength *I*. At 25°C, the constant A takes the value 0.5085. The temperature dependency of the equilibrium constants K was calculated using either the van't Hoff equation:

$$\frac{-d \ln K}{dT} = \frac{\Delta H_r^o}{R T^2}$$

where ΔH_r^o is the reaction enthalpy, R is the gas constant and T is the absolute temperature, or optionally using an empirical equation in T.

RESULTS AND DISCUSSION

Alkalinity and specific alkalinity in inshore waters along the Alexandria coast

During the period of study, for the samples of seawater and the sites of mixing, alkalinity lies in the range 2.24-5.35 meq/l (Fig. 1, table 1), with an average value of 2.99 ± 0.70 meq/l. Most of the samples (~ 70 %) have alkalinity values in the range 2.24-2.99 meq/l. This range lies within that prevailing for seawater. According to Wong (1988), seawater approximates to a half-molar sodium chloride with a rather uniform alkalinity of about 2-3 meq/l. Concerning spatial variation, based on the average values, two maxima were recorded at stations F and D (4.18 ± 1.42 meq/l and 3.41 ± 0.58 meq/l respectively). Station F is the site of mixing between El-Umum Drain (station K, average alkalinity = 4.59 ± 1.75 meq/l) and El-Mex Bay (station E, average alkalinity = 2.57 ± 0.43 meq/l). However station D is under the effect of Kayet Bey Outfalls. Station J (El-Noubaria Draining Canal affected by seawater intrusion) has also high alkalinity value (3.11 ± 1.24 meq/l). Within the Eastern Harbor region (stations A-C), station B has the highest alkalinity (3.09 ± 0.16 meq/l). The rest stations have more or less the same alkalinity values.

For seawater samples (stations A-E and G, H), specific alkalinity varied within a narrow range (0.104-0.192) (table 1). The average value was 0.133 ± 0.017. This value is close to the value of 0.126 that is adopted by Morcos (1970) after Koczy (1965) as an average value for all water masses. The ranges of specific alkalinity are higher at the El-Umum /Bay of Mex site of mixing (station F, 0.499-0.739) than at the El-Noubaria/sea water site mixing (station I, 0.180-0.338). El-Umum Drain has the largest outflow of all the west delta drainage canals (about 10 x $10^6 m^3 day^{-1}$, personal communication).

Table (1): Ranges and averages of alkalinity and specific alkalinity as well as ranges of salinity, temperature and pH values in seawater samples and in the surrounding region.

Seashore		Alkalinty (meq/l)		Specific alkalinty		Salinty**	pH**	Temperature**(°C)
zone		Range	Mean ± SD	Range	Mean ± SD	Range	Range	Range
E.H.	A	2.80 – 2.99	2.89 ± 0.10	0.122 – 0.144	0.133± 0.011	36.232 – 41.081	8.31 – 8.55	16.0 – 29.0
	B	2.95 – 3.26	3.09 ± 0.16	0.137 – 0.150	0.142± 0.007	36.232 – 39.375	8.09 – 8.50	18.4 – 29.0
	C	2.80 – 2.90	2.86 ± 0.05	0.129 – 0.136	0.133± 0.004	36.394 – 38.974	8.26 – 8.54	17.0 – 29.0
Kayet Bey	D	2.99 – 4.08	3.41 ± 0.58	0.135 – 0.192	0.159± 0.030	37.459 – 38.217	7.64 – 8.08	16.0 – 29.0
El-Mex Bay	E	2.29 – 3.07	2.57 ± 0.43	0.122 – 0.136	0.129± 0.007	32.998 – 38.777	8.25 – 8.28	17.7 – 30.3
	F	2.60 – 5.35	4.18 ± 1.42	0.499 – 0.739	0.615± 0.120	9.600 – 14.410	7.77 – 8.00	16.0 – 29.6
	K (source)	2.75 – 6.23	4.59 ± 1.75	1.096 – 2.722	2.119± 0.890	4.150 – 4.670	7.63 – 7.67	16.0 – 29.5
El-Kilo 21	G	2.24 – 2.88	2.50 ± 0.34	0.104 – 0.128	0.117± 0.012	37.854 – 39.821	8.13 – 8.24	17.6 – 30.5
	H	2.45 – 2.93	2.70 ± 0.24	0.114 – 0.128	0.121± 0.007	38.974 – 40.345	8.17 – 8.35	18.1 – 30.5
	I	2.34 – 3.30	2.70 ± 0.53	0.180 – 0.338	0.238± 0.087	14.442 – 33.119	8.07 – 8.25	17.6 – 30.5
	J (source)	2.34 – 4.53	3.11 ± 1.24	0.404 – 0.817	0.546± 0.235	10.570 – 13.990	8.01 – 8.38	17.6 – 30.4

** values were taken from Youssef. and Lees-Gayed (in preparation)

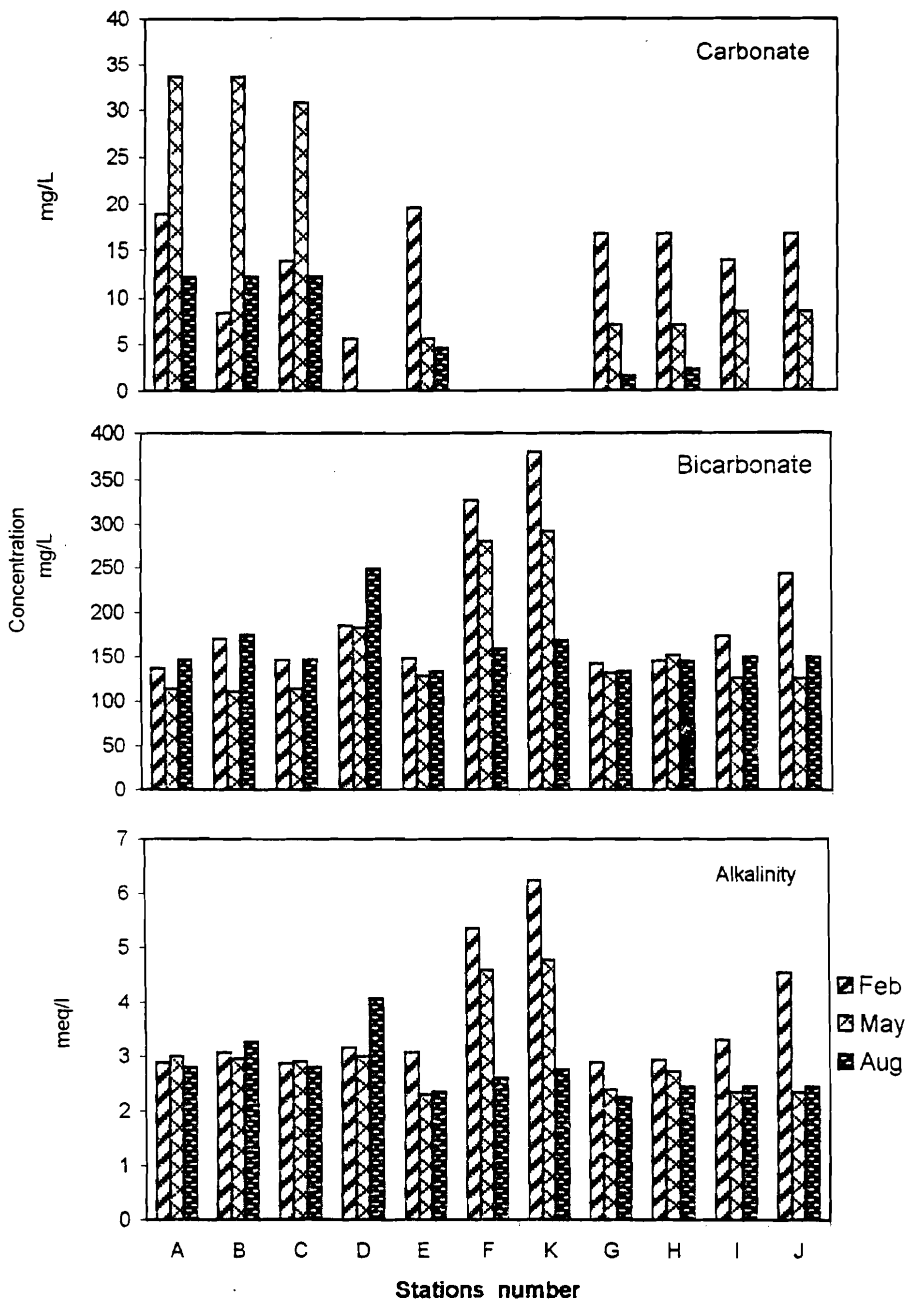


Fig. (1): Concentration of carbonate, bicarbonate and alkalinity in the samples of seawater and the sites of mixing and in El-Umum Drain.

In general, the distribution of alkalinity and specific alkalinity in the studied samples were affected greatly by the discharged drainage waters, which have the highest levels of alkalinity and specific alkalinity (as an example, station K (table 1)). According to Grasshoff (1975), alkalinity increasing in the surface water may be caused partially from the large amounts of fresh water discharges. Indeed, salinity was inversely correlated with alkalinity and with specific alkalinity ($r = -0.40$ and -0.91, respectively, $n= 27$, $p<0.05$). Tayel *et al.* (1996) and Dowidar *et al.* (1987) found the same correlation between the total alkalinity and salinity of the waters of Mex and Dekhaila regions and of the Eastern–Harbour region respectively.

Alkalinity and specific alkalinity in Lake Edku

Throughout the period of study, alkalinity reached a peak of 7.23 meq/l at station 1(Feb 2000) and a minimum of 3.40 meq/l at station 4 (May 2000) (Fig. 2 and table 2). For 70 % of the samples, the alkalinity varied within a limited range (4.05-5.86 meq/l). This range, as expected, exceeded that of seawater samples. During the last decades the conditions in the lake water have been deteriorated rapidly and extensively due to various human impacts. However, unexpectedly, the range of alkalinity during the present study (3.40-7.23 meq/l) is more or less near the range (3.7-6.2 meq/l) determined by Samaan (1974) during 1969-1970. According to Vanloon and Duffy (2000) who made a sensitivity classification of lakes expressed by various measures of alkalinity (table 3), Lake Edku (> 0.4 meq/l alkalinity) may be classified as a low sensitive lake.

The specific alkalinity for Lake Edku fluctuated between 2.74 and 13.29 (Table 2). Based on the averages values, the results show an obvious trend of regional variation. Thus the maximum values of 12.737± 0.580 and 11.822 ± 0.661 were recorded at stations 7 and 8 respectively. Station 7 and to a lesser degree station 8 receive drainage water from El-Khairy Drain in which the specific alkalinity lies in the range 8.233-11.968. Generally, throughout the investigated period, Lake Edku and its connected drains (El-Khairy and Barsiek) have nearly the same specific alkalinity average values (8.780 ± 2.697 and 8.355 ± 2.566 respectively). This indicates that the specific alkalinity may be related to the type of water mixing at the different parts of the lake.

Table (2): Ranges and averages of alkalinity and specific alkalinity as well as ranges of total dissolved solids, pH and temperature values in Lake Edku and in its connected drains.

Stations	Alkalinty (meq/l)		Specific alkalinty		TDS* (mg/l)	pH*	Temperature*(°C)
	Range	Mean ± SD	Range	Mean ± SD	Range	Range	Range
1	4.93 - 7.23	5.96 ± 1.17	2.931 - 7.946	6.230 ± 2.858	1990 - 5940	8.60 - 8.74	17.4 - 26.0
2	3.86 - 6.19	5.09 ± 1.17	6.714 - 10.315	8.614 ± 1.809	1700 - 2310	8.28 - 8.90	15.3 - 26.0
3	4.37 - 5.16	4.67 ± 0.43	7.918 - 8.755	8.323 ± 0.419	1750 - 2270	8.84 - 9.36	16.6 - 26.5
4	3.40 - 5.58	4.45 ± 1.10	8.084 - 10.394	8.888 ± 2.666	1720 - 2440	8.87 - 9.20	15.7 - 26.5
5	4.05 - 6.67	5.17 ± 1.35	4.935 - 8.080	6.393 ± 1.470	1920 - 3280	8.06 - 8.68	16.1 - 26.5
6	4.23 - 7.02	5.47 ± 1.42	5.069 - 10.755	8.567 ± 3.061	1520 - 3240	7.30 - 7.99	16.4 - 26.5
7	5.12 - 5.86	5.49 ± 0.37	12.133 - *13.289*	12.737 ± 0.580	1320 - 1580	7.33 - 8.28	17.3 - 26.5
8	5.12 - 6.33	5.61 ± 0.63	11.156 - 12.479	11.822 ± 0.661	1310 - 2160	7.82 - 8.97	16.8 - 26.5
9	4.98 - 6.37	5.59 ± 0.71	9.048 - 10.880	10.183 ± 0.991	1400 - 2730	8.04 - 8.90	16.4 - 26.5
10	4.81 - 6.05	5.31 ± 0.65	2.735 - 7.740	6.040 ± 2.862	1530 - 4290	7.78 - 8.65	16.5 - 26.5
El-khairy (source)	3.91 - 5.67	4.79 ± 1.25	8.223 - 11.968	10.095 ± 2.649	2300 - 2360	7.49 - 7.65	15.7 - 25.7
Barsik (source)	2.98 - 5.72	4.35 ± 1.94	6.057 - 7.173	6.615 ± 0.789	1330 - 1520	7.94 - 8.09	16.6 - 25.6

* Values were taken from Abbas *et al (in press)*

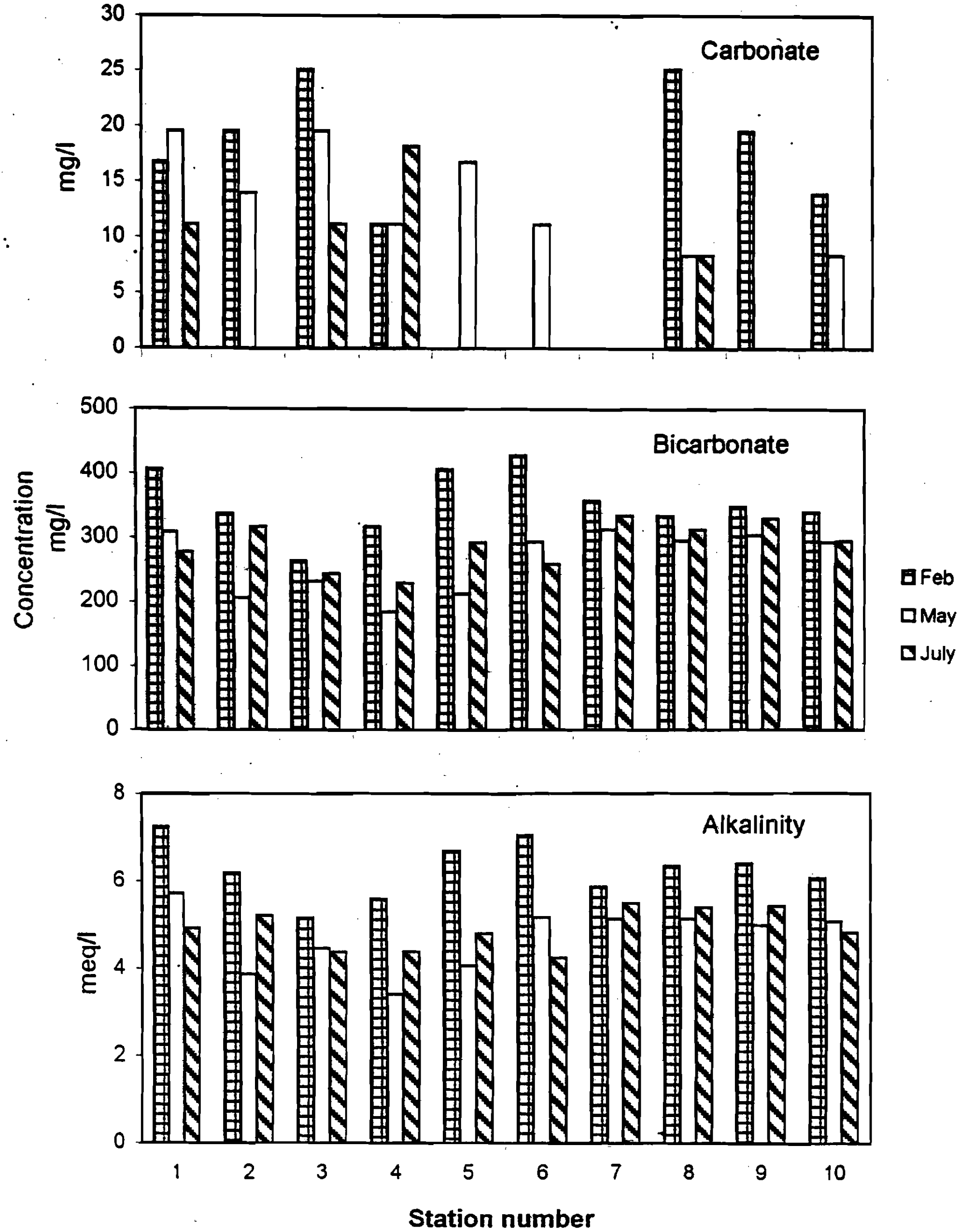


Fig. (2): Concentration of carbonate, bicarbonate and alkalinity in the samples of Lake Edku.

During the period of study, specific alkalinity is inversely correlated with total dissolved solids ($r = -0.66$, $n=30$, $p<0.05$). However, the reversed relationship is observed between the total dissolved solids and alkalinity ($r = 0.50$, $n=30$, $p<0.05$). Similar observations were reported by Mahlis *et al* (1970) for Lake Mariut.

The ranges of alkalinity and specific alkalinity for Lake Edku (3.40-7.23 meq/l and 2.74-13.29 respectively, Table 2) during the present study are noticeably lower than those observed for the Lake proper basin of Lake Mariut (3.56-10.58 meq/l and 4.79-16.41 respectively), but they are significantly higher than those measured for the north-western basin of Lake Mariut (3.77-4.69 meq/l and 1.13-1.62 respectively) (Youssef, 1999).

Table 3: Sensitivity classification of lakes expressed by various measures of alkalinity*.

Sensitivity	meq/l
High	< 0.2
Moderate	0.2-0.4
Low	> 0.4

* After Vanloon and Duffy (2000)

<u>*Carbonate and bicarbonate levels in the investigated regions*</u>

Despite the variety of source water, bicarbonate is the dominant species of the CO_2 system. It contributed 88 %, 94 %, and 100 % of the total alkalinity of the samples of seawater, Lake Edku and drain water respectively (Fig.3).

For the samples of seawater and the sites of mixing between the drains (El-Umum Drain and El-Noubaria Canal) and the seawater, the levels of bicarbonate exhibited a wide range (111.30-326.28 mg/l), with an average value of 160.91 ± 49.85 mg/l (Fig.1). This may be due to the different in salinity as a result of mixing of sea water with drains. Indeed, there is a significant negative correlation between the concentration of bicarbonate and salinity ($r = -0.51$, $n=27$, $p<0.05$). On the other hand, carbonate levels (see below) are positively correlated with salinity ($r = 0.42$, $n=27$, $p<0.05$). This indicates that the concentration of bicarbonate increases with increasing drain water of fresh origin. However, the reverse situation is true for carbonate. Rai (1974) emphasized the effect of sewage effluents in increasing bicarbonate concentrations.

For Lake Edku, the concentrations of bicarbonate (Fig. 2) varied between 184.42 and 428.42 mg/l, with an average value of 302.56 ± 58.24. This range is clearly higher than the ranges observed for the North-Western basins of Lake Maruit (168.3-260.9 mg/l, Youssef (1999)) and Lake Manzalah (18-270 mg/l, Abdel-Moati (1985)). However, the range found in the present investigation is quite near the range (70-462 mg/l) determined by Abdel-Moati (1985) for the Lake proper of Lake Manzalah, but it is significantly lower than that for the Lake proper of Lake Mariut (253.2-645.2 mg/l, Youssef (1999)).

For Lake Edku samples (Fig. 2) the highest carbonate value does not exceed 25.12 mg/l., whereas for seawater region (Fig. 1) the maximum concentration reached 33.68 mg/l. For both Lake Edku and seawater samples, a strong positive correlation was observed between the concentration of carbonate and pH of the water ($r = 0.78$, $n=30$ and $r = 0.68$, $n=27$ respectively). Indeed, CO_3^{2-} dominates in high pH conditions (Horne and Goldman (1994)). Throughout the period of study, the water of El-Khairy, Barsiek and El-Umum Drains as well as their surrounding regions (station 7 and station F) were completely devoid of carbonate. This may be attributed to the effect of drainage water which is poor in dissolved oxygen and has low pH values. Indeed, during the same period of study, El-Khairy and Barsiek drains had the lowest levels of both dissolved oxygen and pH values (1.39-2.70 mg/l and 7.49-8.09 respectively) compared with those in Lake Edku water (Abbas *et al.*, in press). Likewise, Gharib and Soliman (1998) recorded the lowest values of pH in drains sector of Lake Edku. Depletion of carbonate at the sites of disposal of drains water was also observed by Abdel-Moati (1985) for Lake Manzallah. This lake is oxygenated except for the area localized around the inlet of Bahr El-Bakar Drain, which is always anoxic (8.1-20.64 ml/l, Dowider *et al* 1984). Similarly several stations in the Lake proper basin of Lake Mariut (a heavily polluted basin) were devoid of carbonate (Youssef, 1999).

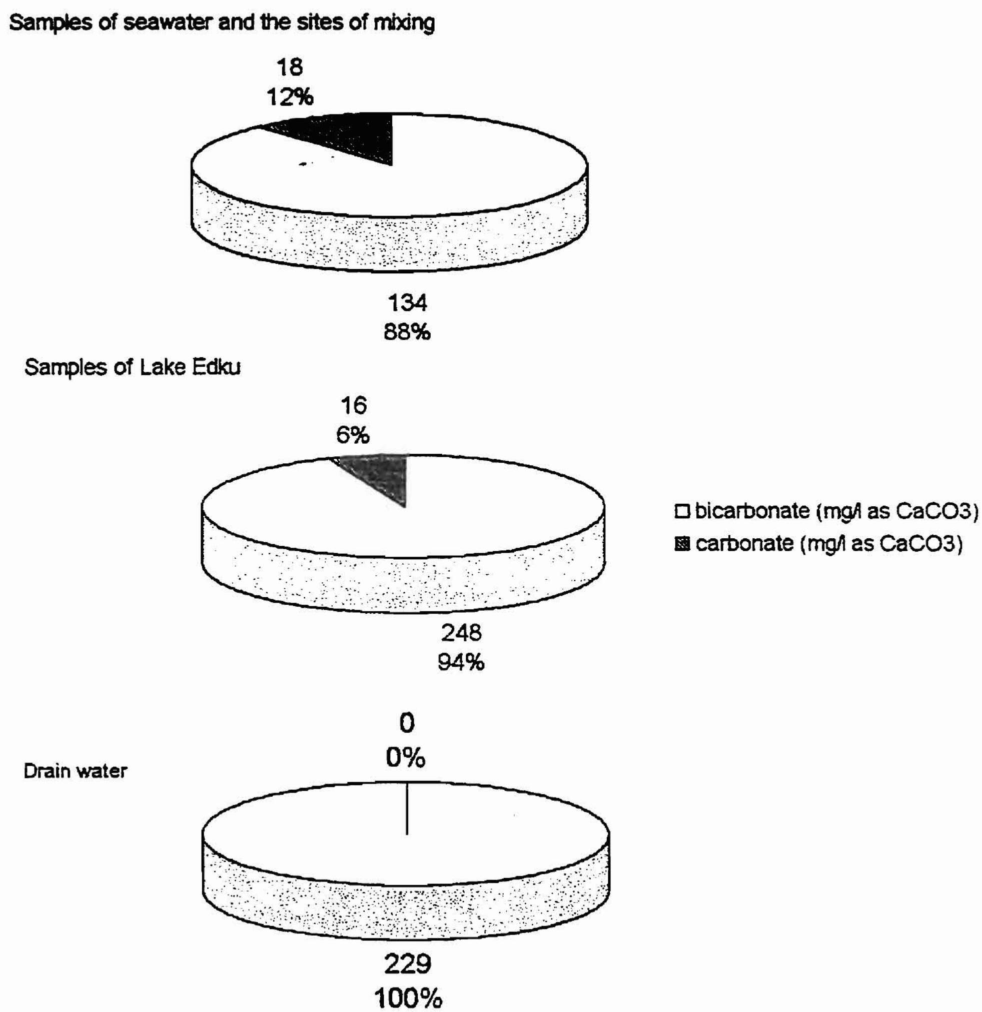


Fig. 3 Average values of carbonate and bicarbonate alkalinities and their percentages to total alkalinity in the studied samples

Modelling studies

Since bicarbonate is the dominant forms of the CO_2 system, modelling studies are carried out to determine the different species of bicarbonate in the studied regions. The main calculated species are the free ion HCO_3^- and the ion pairs $NaHCO_3^0$ and $MgHCO_3^+$ (table 4). In all the environments studied, the free ion HCO_3^- is the dominant species. Its percentage ranged from 79.04 % in seawater samples to 97.90 % in the brackish water of El-Khairy Drain. The percentages of the ion pairs $NaHCO_3^0$ (0.48 to 9.98 %) and $MgHCO_3^+$ (1.06 to 8.76 %) are nearly close to each other. The highest percentages are for the seawater samples. However, El-khairy Drain has the lowest percentages. The sites of mixing have moderate percentages of both the free ion and the ion pairs. It was observed that seawater samples have the lowest free ion percentage and the highest percentages of the ion pairs. This is due to the presence of high concentrations of sodium and in seawater magnesium samples compared with those in the other studied environments.

There is a great consistency in the speciation of bicarbonate in Lake Edku and in its connected Drains (El-Khairy and Barsiek). The percentages of the $MgHCO_3^-$ ion pair exceeded those of $NaHCO_3^0$ in the water of Lake Edku and its connected drains. However, in seawater the percentage of $NaHCO_3^0$ is slightly higher than that of $MgHCO_3^+$. These results can be explained by considering the values of formation constants of the ion pairs (log k_{MgHCO3}= 1.061, log k_{NaHCO3} = -0.25. Martell *et al* 1997) and the relative concentrations of sodium and magnesium in the various environments (table 5).

CONCLUSIONS

Distribution of total alkalinity, specific alkalinity, carbonate and bicarbonate in some Egyptian aquatic environments subjected to land-runoff were carried out. It was concluded that:

1) Despite the variety of source water, bicarbonate is the dominant species of the CO_2 system.
2) The maximum average ratios of specific alkalinity were recorded at regions affected by the drainage water.
3) Computer equilibrium modeling for the bicarbonate species show that the free ion HCO_3^- and the ion pairs $NaHCO_3^0$ and $MgHCO_3^+$are the main forming bicarbonate species.

Table (4): Means of the percentages* of HCO_3^- -containing species obtained from modelling of different aquatic environments.

Species / Aquatic environments	HCO_3^-	Na HCO_3^0	$MgHCO_3^+$
-Sea water samples (n=21)	79.04±0.67	9.98±0.51	8.76±0.24
- Sites of mixing.			
-Site of mixing between the Noubaria Draining Canal and the sea water of El-kilo 21(n=3)	84.55±3.53	6.64±2.07	6.62±1.84
	89.24±1.12	4.19±0.49	4.32±0.56
-Noubaria Draining Canal before mixing with the sea (n=3)			
	89.79±0.46	4.02±0.27	4.58±0.59
-Site of mixing between El-Umum Drain and the sea water of El-Mex Bay (n=3)			
-Lakes	97.32±0.56	0.69±0.17	1.37±0.31
-Lake Edku (n=30)			
-Drains	93.81±0.91	2.18±0.68	2.40±0.13
-El- Umum Drain (n=3)	97.90±0.54	0.48±0.06	1.06±0.16
-El-Khairy Drain(n=2)	97.22±0.62	0.64±0.15	1.39±0.12
-Barsiek Drain(n=2)			

n= number of samples * percentages of main species

Table (5): Average levels (mg/l)of bicarbonate, sodium and magnesium in the studied regions

	bicarbonate	sodium*	magnesium*	Na/Mg ratio
–seawater samples	157.21	11556.82	1366.32	8.46
Site of mixing between the Noubaria Draining Canal and the sea water of km21	*149.28*	*7200.00*	*915.16*	*7.87*
–Noubaria Draining Canal before mixing with the sea.	172.45	3889.33	472.72	8.23
Site of mixing between El-Umum Drain and the sea water of El-Mex Bay.	*254.82*	*3630.00*	*469.69*	*7.73*
–Lake Edku	302.56	417.73	71.91	5.81
–El-Umum Drain	279.71	1630.33	175.75	9.28
–El-Khairy Drain	292.24	273.57	45.82	5.97
–Barsiek Drain	265.28	389.05	74.55	5.22

* values were taken from Youssef. and Lees-Gayed (in preparation)

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