

TRACE METALS ACCUMULATION IN MOLLUSCAN SHELLS FROM DAMIETTA NILE BRANCH SEDIMENTS, EGYPT

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ABSTRACT

The dissolved heavy metals (As, Ba, Cd, Cr, Co, Cu, Fe, Mn, Mo, Ni, Pb, Se, Sr, V and Zn) in molluscan shells from Damietta Nile branch sediments were estimated in three zones during April – June 2004. The average values ($\text{ppm} \times 10^{-3}$) were 0.63, 0.3, 0.01, 0.05, 0.13, 2.32, 3.33, 1.97, 0.19, 0.22, 0.26, 0.5, 2.51, 0.051 and 2.21, respectively.

Results indicate that levels of heavy metals increased northwards and in neopionic (recent) stages and were positively correlated with salinity, chlorinity, alkalinity, dissolved oxygen and calcite mineral and negatively correlated with pH values and aragonite mineral. Certain heavy metals such as As, Co, Cu, Fe, Mn, Mo and Pb were more abundant in bivalve shells (i.e. high aragonite), while Ba, Cd, Cr, Ni, Pb, Sr, V and Zn were more abundant in gastropod shells (i.e. high calcite).

INTRODUCTION

Under natural conditions, the most important inputs of metals to aquatic regions are the mechanical weathering of rocks (Bryan, 1984).

There are numerous types of pollutants in the aquatic environment such organic matter, major and trace metals contribute to both natural and anthropogenic sources including storm dust-fall, erosion or crustal weathering and decomposition of biota in water, whereas the anthropogenic sources include sewage, industrial and mobile wastes as shipwrecks and dumping of war materials (Al-Saad 1995; Lotfy 1997; Ansari *et al.* 1999).

The direct discharge of wastes containing trace metals to the environment, tends to increase in water column, while sediments act as archive for pollutants (Barnes *et al.*, 1984; Barcellos *et al.*, 1988), and may quantitatively and qualitatively alter the natural biochemical cycle

(Grimanis *et al.*, 1978). As a part of the aquatic environment, recent shells accumulate trace metals and act as indicators of pollution (Roy and Crawford, 1984; Kilby and Batley, 1993; Lotfy, 1997, 2000 and 2001).

A few reports of heavy metals concentrations in freshwater molluscan shells have been documented. Several studies on the pollution of molluscan shells by trace metals (Turikian and Armstrong, 1960; Dodd, 1963; FAO, 1964; Lotfy, 1997, 2000, 2001, 2002 and 2003), were taken into consideration.

The Damietta Nile branch extends north of Al- Kanater Barrage for about 218 km along the eastern boundary of the Nile Delta Egypt and it leads finally to the Mediterranean Sea after Faraskour dam (Fig. 1). It varies in width from 180 and 250m. The branch represents a shallow water stream with a depth fluctuating between 4.5 and 16m. The bottom sediments range from gravelly sand and sand units in the beach and southwards to gravelly muddy sand and muddy sand units in the deep zone and northwards (Lotfy, 1997).

This study aimed to determine the distribution of trace metals in some molluscan shells to illustrate the relation between their accumulation, metabolism, secondary alteration, environmental parameters and the sources of these pollution.

MATERIALS AND METHODS

A total of 50 samples were collected from recent bottom sediments, representing the area of Damietta Nile branch and distributed along ten stations (Fig. 1). Each station was represented by five samples (i.e. two from eastern, two from western beach and one from the mid- stream).

The bottom samples were collected by a Peterson dredge grap sampler from different locations during April- June, 2004.

Water samples were collected from different locations in polyethylene bottles using water sampler. Water temperatures were recorded using thermometer. Dissolved oxygen and salinity were determined according the methods described by Strickland and Parsons (1972). The pH values were measured by a pocket pH meter. The total alkalinity was determined by titration versus standard HCl, using methyl orange indicator. The total chlorides were determined following Mohr's titration.

The shells of the bivalve *Anodonta implicata* (nepionic and adult stages and the gastropod *Bellamiya unicolor* (nepionic and adult stages were picked up from Damietta Nile branch sediments.

The determination of trace metals As, Ba, Cd, Cr, Co, Cu, Fe, Mn, Mo, Ni, Pb, Se, Sr, V and Zn in the studied species was done using the Inductivity Coupled Plasma (ICP) in the Central Laboratory for Environmental Quality Monitoring (CLEQM), National Water Research Center, Kanater, Kalyubeya, Egypt.

RESULTS AND DISCUSSION

Environmental Parameters of Water

Table (1) and Fig. (2) illustrate several important factors concerning the spatial distribution of pH value, oxygen content, chlorinity, alkalinity in Damietta Nile branch water, and the distribution of carbonate mineral phases of bivalve and gastropod shells in different zones of study area by Lotfy (1997).

The Damietta Nile branch water temperature was subjected to both diurnal and seasonal variations (Lotfy, 1997). The monthly variation of the water temperature ranged between 14.5°C (January) and 32°C (June). The pH values of water ranged from 7.5 at Damietta to Ras El-Bar area to 8.11 at El-Kanater to El- Mansoura; it lies mostly on the alkaline side. The salinity of water concentrated in the Estuary of Damietta ranged between 35 and 42.51‰. the content of chlorinity ranged from 40.6 mg/l at El-Kanater (southern region) to 79.5 mg/l northwards. Then the high concentration was observed at Damietta Estuary (close to the Mediterranean Sea). The dissolved oxygen varied at different sites and the water was completely oxygenated from surface to bottom. It's content ranged from 7.54 mg/l at the southern area to 9.51 mg/l at Damietta Estuary (Fig. 2). Total alkalinity of Nile water attained a highest value northwards with 196.3 mg/l (Fig. 2), then declined to minimum value southwards with an average of 164.5 mg/l.

Lotfy (1997) proved that the carbonate mineral phases of bivalve and gastropod shells consist mainly of aragonite and calcite. The aragonite decreased northwards, while the calcite increased. In gastropod shells, the maximum value of aragonite was recorded southwards reaching as high as 95% and the minimum value of aragonite occurred northwards attaining 25% (Fig. 2 and Table 1), while the calcite ranged between 5% (southwards) and 75% (northwards). The aragonite contents in bivalve shells ranged from 95.5% (southwards) to 75% (northwards) and the calcite varied between 3.5% (southwards) and 25% (northwards).

Trace Metals in Bivalve Shells

Concentrations of the trace metals arsenic (As), barium (Ba), cadmium (Cd), chromium (Cr), cobalt (Co), copper (Cu), iron (Fe), manganese (Mn), molybdenum (Mo), nickel (Ni), lead (Pb), selenium (Se), strontium (Sr), vanadium (V) and zinc (Zn) in the bivalve *Anodonta implicata* shell are listed in Table (2) and presented in Fig. (3). For As, Ba, Cd, Cr and Co, the mean levels due to *Anodonta implicate* shells ranged between 0.61- 1.33, 0.04- 0.31, 0.001- 0.013, 0.02- 0.096 and 0.01- 0.075 ppm $\times 10^{-3}$, respectively. In the northern zone (close to the Damietta city), the average contents of As and Cd showed a high content in nepionic stage, while Ba in the adult stage. In the southern region (near to the industrial region in Cairo), Co and Cr showed maximum values in nepionic stage. Meanwhile, the minimum values of As, Ba and Cd appeared in southern zone in adult stage, and the lowest value of Co showed in nepionic stage (northwards). Finally, the lowest value of Cr was recorded in mid- stream in the adult stage.

According to the distribution of Cu, Fe, Mn, Mo and Ni (Table 2 and Fig. 3), the northern zone showed a maximum values of Cu, Fe, Mn and Mo in nepionic stage and Ni in the adult stage (7.71, 5.99, 3.41, 0.5 and 0.09 ppm $\times 10^{-3}$, respectively). Towards the southern region, the values of Cu, Mn, Mo and Ni became very low (0.16, 0.98, 0.12 and 0.018 ppm $\times 10^{-3}$, respectively) in the adult stage, while the lowest value of Fe (0.78 ppm $\times 10^{-3}$) was recorded in the adult stage (northwards).

Generally, the trace metal levels in *Anodonta implicata* shells were more abundant in nepionic stage (northwards).

As shown in Table (2) and Fig. (3), the levels of Pb, Se, Sr, V and Zn ranged between 0.19- 0.61, 0.18- 0.87, 0.16- 1.5, 0.017- 0.09 and 0.52- 2.68 ppm $\times 10^{-3}$, respectively. Changes in total levels of Pb, Se, Sr and Zn in *Anodonta implicate* shells from Damietta Nile branch sediments attained the highest level in nepionic stage in northern area with 0.61, 0.87, 1.5 and 2.68 ppm $\times 10^{-3}$, respectively, while the highest value of V was attained in the adult stage (northwards) with 0.09 ppm $\times 10^{-3}$. Their levels became very low in adult stage southwards.

Enrichment Factors (E.F.) for the accumulation of trace metals in the different stages of bivalve shells can be derived using the following expression:

E.F. =	mean concentration of trace metals in the nepionic stage
	mean concentration of trace metals in the adult stage

From Table (4) with the exception of the southern sector(close to the industrial region at Greater Cairo), the E.F. for As, Ba, Cd, Co, Cr, Cu, Fe, Mn, Mo, Ni, Pb, Se, Sr, V and Zn were greater than 1.1, 7, 9, 1.23, 5.29, 43, 6.28, 1.57, 1.33, 4.33, 3.05, 4.67, 4.25, 4, and 4.64, respectively. These factors indicated that the nepionic stage of bivalve shells were more highly polluted southwards (i.e. highly correlated with aragonite content).

Meanwhile, in the middle sector(close to El-Mansoura city), the E.F. for As, Ba, Cd, Cr, Cu, Fe, Mn, Mo, Pb, Se, and Zn were greater than 1.17, 1.14, 6, 5, 32.4, 1.25, 2.21, 2.05, 2.62, 1.21 and 1.45, respectively, while Co, Ni and V levels exhibited low enrichment in nepionic stage.

Finally, in the northern sector (near to Damietta city), the E,F, for As, Ba, Cd, Cr, Cu, Fe, Mn, Mo, Pb, Se, Sr and V contents were greater than 1.08, 4.33, 3.5, 8.04, 7.68, 1.54, 2.38, 2.18, 2.72, 1.44 and 1.04, respectively, while Ba, Co, Ni and V values in nepionic stage exhibited low enrichment.

Trace Metals in Gastropod Shells

Levels of trace metals As, Ba, Cd, Co, Cr, Cu, Fe, Mn, Mo, Ni, Pb, Se, Sr, V and Zn in the gastropod *Bellamiya unicolor* shell are listed in Table (3) and presented in Fig. (4). Appreciable difference could be observed in the abundance of the elements in all stations and in nepionic and adult stages with relatively high levels for most metals at northern region. Their levels ranged between 0.16- 0.6, 0.26- 0.63, 0.01- 0.019, 0.013- 0.04, 0.018- 0.56, 0.12- 1.8, 0.88- 5.7, 1.58- 2.28, 0.08- 0.19, 0.004- 1.04, 0.12- 0.23, 0.08- 1.3, 0.03- 0.36, 1.48- 6.9, 0.018- 0.16, and 0.94- 3.28 ppm $\times 10^{-3}$, respectively.

Concerning the distribution of As, Ba, Cd, Co and Cr (Fig. 4), the highest levels of As, Ba and Cr were attained in the northern side in nepionic stage, while Co in the adult stage, finally the highest value of Cd occurred in the southern region in nepionic stage. Generally, their levels were less abundant in southern zone, while Cd value decreased northwards.

However, the gradient of Cu, Fe, Mn, Mo and Ni levels variation in *Bellamiya unicolor* shells are shown in Fig. 4. The maximum value of Cu, Fe and Mo (1.8, 5.7 and 0.19 ppm $\times 10^{-3}$, respectively) was observed in adult stage (northwards), while Ni reached a highest level (2.28 ppm $\times 10^{-3}$) in nepionic stage (southwards).

Levels of Pb, Se, Sr, V and Zn in shells of *Bellamiya unicolor* showed a general increase northwards, where high levels of Pb, Sr and Zn

(0.23, 6.9 and 3.28 ppm $\times 10^{-3}$, respectively) were recorded in nepionic stage, while high values of Se and V (1.3 and 0.16 ppm $\times 10^{-3}$, respectively) were measured in adult stage and decreased southwards, while Se decreased towards the middle zone.

From Table (4), with the exception of the southern sector, the E.F. for Ba, Cd, Cr, Mn, Ni, Pb, Sr and Zn in gastropod shells were greater than 1.46, 1.46, 7.78, 1.44, 5.68, 1.11, 5.33 and 2.68, respectively, while As, Co, Cu, Mo, Se and V exhibited low enrichment in nepionic stage.

In the mid- stream region, most of elements exhibited high values, while Co, Cu, Fe, Mo, Se and V exhibited low values in nepionic stage.

From Table (4), in northern sector, the E.F. for As, Ba, Cr, Mn, Ni, Pb, Se and Zn were greater than 2.4, 1.91, 2.67, 1.15, 1.16, 1.1, 1.92 and 1.3, while Cd, Co, Cu, Fe, Mo, Se and V exhibited low value. generally, most of elements in the gastropod shells are more abundant in nepionic stage (southwards).

The relative order of abundance of elements in the molluscan shells was as follows: As, Co, Fe, Mn, Mo and Pb that were more associated in bivalve shells, while Ba, Cd, Cr, Ni, Sr, V and Zn were more associated in gastropod shells, and Se have a uniform distribution in molluscan shells.

Trace Metals Distribution and Environmental Factors

From Figs. (2, 3 and 4), the relationship matrix among trace metals levels in molluscan shells and minerals composition of shells and environmental parameters of water demonstrates the following: the combined values show strong pathetic and antipathetic relations; chlorinity, dissolved oxygen, alkalinity, salinity, calcite, most of elements in bivalve shells (except Co, Fe and V) and all elements in gastropod shells were positively correlated with each other and increased northwards and conversely, they exhibited negative relationships with pH value, aragonite and Co in bivalve shells, while Fe and V had a uniform distribution in all zones. So, most of elements in molluscan shells except Co, Fe and V were more associated with calcite mineral, while Co were more associated with aragonite. This depends on metabolism, activity of organism, early diagenesis and environmental factors.

Thus, as expected most of trace metals are incorporated into carbonate mineral phases (Batley, 1987; Fernades, 1997; lotfy, 1997, 2000, 2001, 2002 and 2003).

The relative order of abundance of elements in the bivalve shells was as follows: Cu > Fe > Zn > Mn > As > Sr > Se > Pb > Mo > Ba > Co > Ni > V >

Cr> Cd and most of trace metals increased northwards. In the gastropod shells, their abundance was as follows: Sr> Fe> Zn> Mn> Cu> Se> Ni> Ba> As> Cr> Pb> Mo> V> Co> Cd and all elements increase northwards.

The results also proved that As, Co, Cu, Fe, Mn and Pb are more abundant in bivalve shells, while Ba, Cd, Cr, Ni, Sr, V and Zn are more abundant in gastropod shells, but Se has a uniform distribution.

The zonal distribution of trace metals in molluscan shells by using E.F. proved that most elements exhibited high enrichment in nepionic stage northwards and southwards. Then, the main source of pollution is the industrial region in greater Cairo and Damietta cities.

Matching the average values of elements in molluscan shells in Damietta Nile branch sediments with those in carbonate sediments (Turikian and Wedepohl,1961), the results showed that Se is highly enriched in the molluscan shells of Damietta Nile branch. In comparison with the standard shale (Krauskopf and Bird, 1995), results showed that the elements are lowly enriched in the molluscan shells.

Forestner and Muller (1973), Forestner and Wittman (1981) and Forestner (1982) proved that the remobilization of metals from sediments is mainly controlled by the absorbed processes and is mostly caused by four types of chemical changes in water: 1- elevated salt contents; 2- changes in the redox; 3- lowering pH and 4- presence of natural complexion agents, in addition to biochemical processes and activity of organisms.

CONCLUSION

Molluscan shells from Damietta Nile branch sediments are slightly contaminated with heavy metals. In spite of the high concentrations recorded in the northern and southern sector, they do not exceed the International permissible concentrations in carbonate sediments and shale.

From the results obtained, the northern sector was slightly polluted with trace metals due to the effect of the Damietta city wastes, followed by southern sector which is close to the greater Cairo, while in the middle sector, the concentrations of heavy metals in the molluscan shells showed lower values.

The relative order of abundance of elements in the bivalve shells was as follows: Cu> Fe> Zn> Mn> As> Sr> Se> Pb> Mo> Ba> Co> Ni> V> Cr> Cd and most of trace metals increased northwards. In the gastropod shells, their abundance was as follows: Sr> Fe> Zn> Mn> Cu> Se> Ni> Ba> As> Cr> Pb> Mo> V> Co> Cd and all elements increased northwards.

The results proved that As, Co, Cu, Fe, Mn and Pb are more abundant in bivalve shells, while Ba, Cd, Cr, Ni, Sr, V and Zn are more abundant in gastropod shells, but Se has a uniform distribution.

The trace metals distribution in adult and nepionic stages and by depending on the recorded E.F., most elements in bivalve and gastropod shells were more associated with nepionic stage north- and southwards.

The distribution of trace metals in the recent molluscan shells is mainly controlled by some factors as; elevated salt, increasing alkalinity, chlorosity, dissolved oxygen, transformation of aragonite to calcite and lowering pH tend to increasing the absorption processes of elements, in addition to activity of organism and metabolism (I.e. trace metals are more associated in gastropod shells).

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DAMIETTA NILE BRANCH SEDIMENTS

Table(1): The recorded average measurements of some mineralogical and chemical environmental parameters in water and carbonate phases of A-bivalve B-gastropod shells from Damietta Nile branch.

Sediment		--I--		water			
zone	Aragonite%		Calcite%		pH	Alkal. Mg/l	chlorosity Mg/l
	A	B	A	B			
South	96.5	95	3.5	5	8.23	164.5	40.6
Middle	97	91	3	9	8.11	190.3	60.33
north	75	25	25	75	7.5	196.3	79.5

Table(2): Average concentration of trace metals (ppm $\times 10^{-3}$) in shells of *Anodonta implicata* (bivalve A= adult, B= nepionic).

zone	*	As	Ba	Cd	Co	Cr	Cu	Fe	Mn	Mo	Ni	Pb	Se	Sr	V	Zn
south	A	.62	.04	.078	.08	.01	.16	.92	.98	.12	.02	.19	.18	.16	.02	.52
	B	.68	.28	.009	.1	.07	6.88	5.8	1.5	.16	.08	.58	.84	.68	.07	2.3
	Ave	.65	.16	.005	.09	.04	3.52	3.5	1.3	.14	.05	.18	.51	.42	.04	1.6
Mid.	A	.92	.22	.002	.08	.01	.22	.81	1.7	.14	.07	.22	.21	.99	.06	1.7
	B	1.08	.25	.012	.07	.05	7.12	5.8	2.1	.31	.05	.45	.55	1.2	.03	2.5
	Ave	1	.24	.007	.08	.03	3.67	3.3	1.9	.23	.06	.34	.38	1.1	.05	2.1
north	A	1.23	.32	.003	.09	.02	.33	.78	2.2	.21	.09	.28	.32	1.1	.09	2.6
	B	1.33	.16	.013	.02	.07	7.71	6	3.4	.5	.02	.61	.87	1.5	.01	2.7
	Ave	1.28	.24	.008	.07	.05	4.02	3.4	2.8	.37	.05	.45	.6	1.3	.05	2.7
	Tot.	.98	.21	.007	.08	.04	3.74	3.4	2	.25	.05	.32	.5	.93	.05	2.6
**			10	-	.1	11	4	-	110	.4	20	9	.1	610	20	20
***		130	580	.3	19	90	45	472 00	850	2.6	68	20	.4	300	130	95

*= Type of shells **= Standard of carbonate (Turkian and Wedepohl, 1961) all value $\times 10^{-3}$

***=Standard shale (Krauskopt and Bird, 1995) all value $\times 10^{-3}$

Table(3): Average concentration of trace metals(ppm $\times 10^{-3}$) in the gastropod shells of *Bellamiya unicolor* (C= adult, D=nepionic).

zone	*	As	Ba	Cd	Co	Cr	Cu	Fe	Mn	Mo	Ni	Pb	Se	Sr	V	Zn
south	C	.18	.26	.013	.014	.018	.19	3.42	1.58	.11	.019	.18	.18	.148	.03	.94
	D	.16	.38	.019	.012	.014	.12	.88	2.28	.08	.108	.2	.11	5.33	.02	2.7
	Ave	.17	.32	.016	.013	.08	.15	2.15	1.93	.1	.064	.19	.15	3.41	.02	1.8
Mid.	C	.2	.28	.012	.03	.08	1.3	4.11	1.68	.13	.06	.12	.2	2.08	.03	1.7
	D	.3	.41	.015	.018	.21	.9	2.08	2.11	.09	.31	.17	.08	5.05	.02	2.7
	Ave	.25	.35	.014	.024	.15	1.1	3.1	1.9	.11	.19	.15	.14	3.57	.03	2.3
north	C	.25	.33	.01	.04	.21	1.8	5.7	1.81	.19	.9	.21	1.3	3.6	.16	2.5
	D	.6	.63	.009	.022	.56	1.12	3.61	2.08	.11	1.04	.23	1.1	6.9	.07	3.3
	Ave Tot.	.43 .28	.48 .38	.01 .013	.031 .023	.39 .21	1.46 .9	4.66 3.3	1.95 1.93	.15 .12	.97 .39	.22 .19	1.2 .5	5.25 4.08	.12 .06	2.9 2.3
**			10	-	.1	11	4	-	110	.4	20	9	.1	610	20	20
***		130	580	.3	19	90	45	47200	850	2.6	68	20	.4	300	130	95

*= Type of shells **= Standard of carbonate (Turkian and Wedepohl, 1961) all value $\times 10^{-3}$ ***=Standard shale (Krauskopt and Bird, 1995) all value $\times 10^{-3}$

Table(4): Enrichment factors of trace metals in nepionic / adult stage of A-bivalve and B- gastropod shells.

zone	*	As	Ba	Cd	Co	Cr	Cu	Fe	Mn	Mo	Ni	Pb	Se	Sr	V	Zn
south	A	1.1	7	9	1.23	5.29	4.3	6.28	1.5	1.33	4.33	3.05	4.67	4.25	4	4.94
	B	.89	1.46	1.46	.96	7.78	.63	.26	1.44	.73	5.68	1.11	.61	3.6	.67	2.85
Mid.	A	1.17	1.14	6	.89	5	3.4	7.2	1.25	2.21	.73	2.05	2.62	1.21	.05	1.45
	B	1.5	1.46	1.25	.06	2.63	.69	.51	1.26	.69	5.17	1.42	.4	2.43	.67	1.74
north	A	1.08	.52	4.33	.22	3.5	8.04	7.68	1.54	2.38	.22	2.18	2.72	1.44	.08	1.04
	B	2.4	1.91	.9	.55	2.67	.62	.63	1.15	.58	1.16	1.1	.85	1.92	.44	1.3

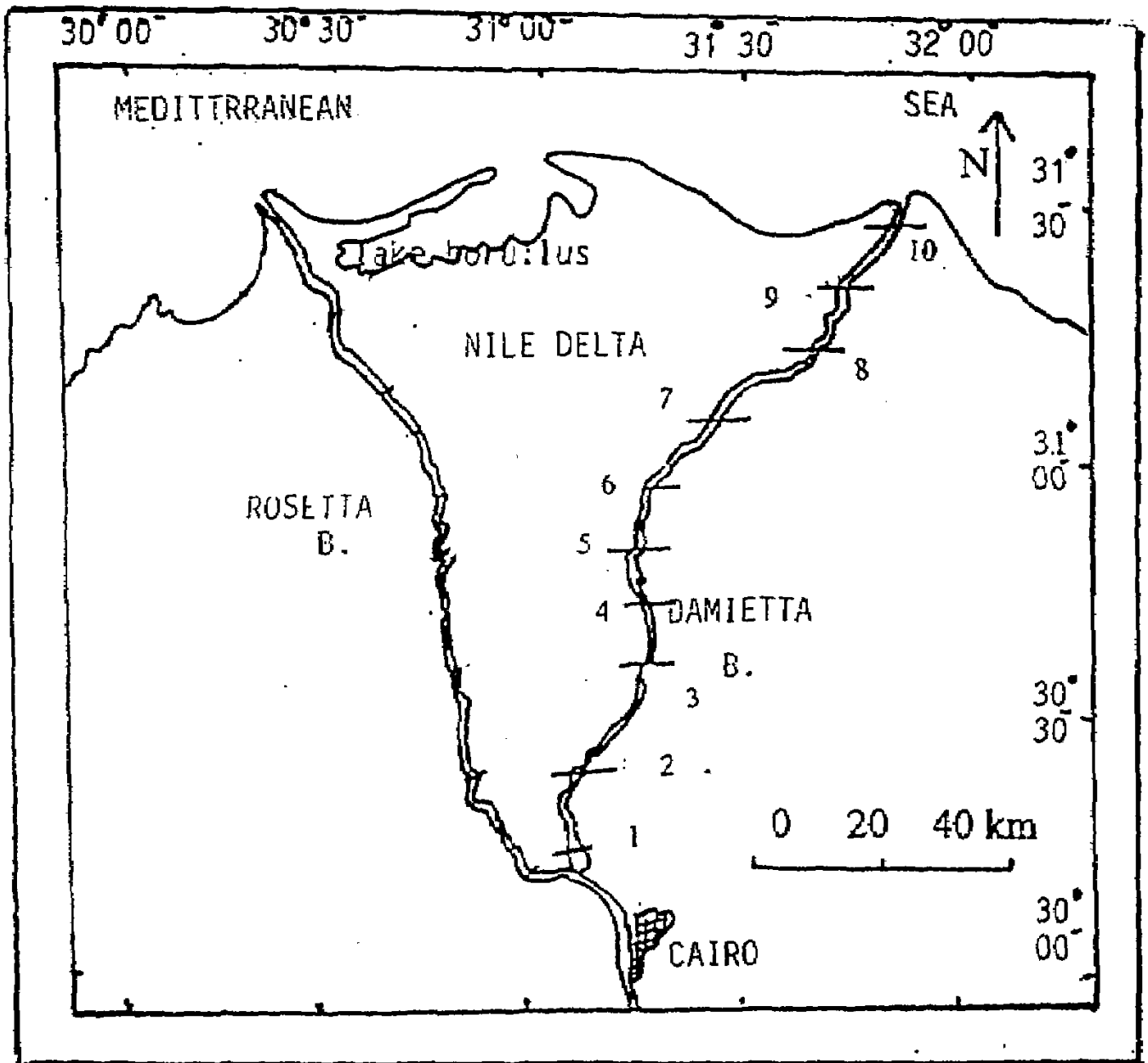
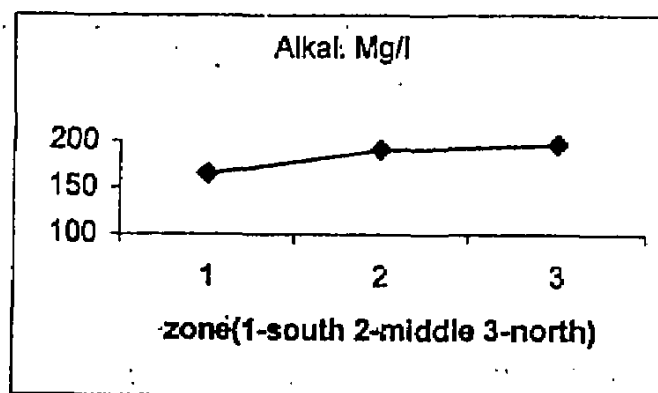
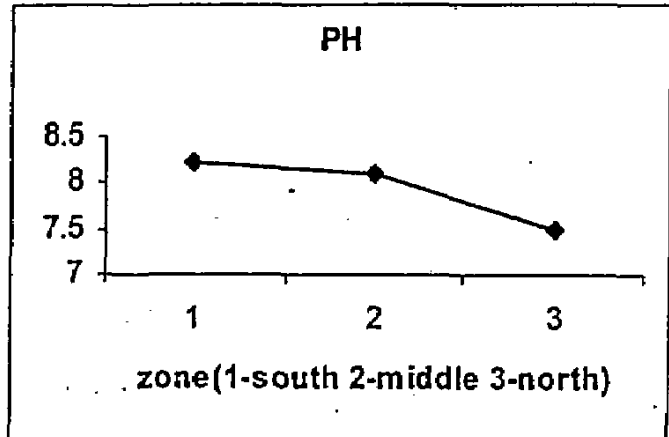
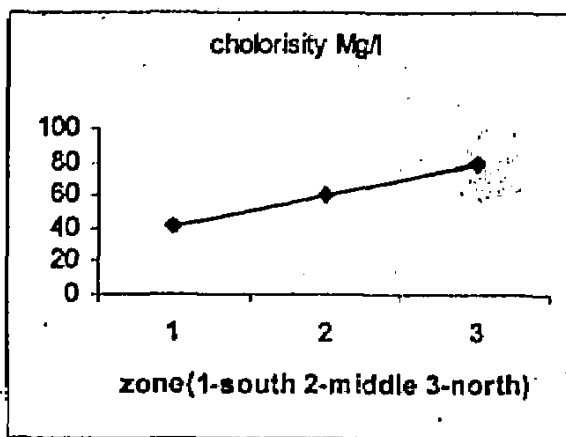
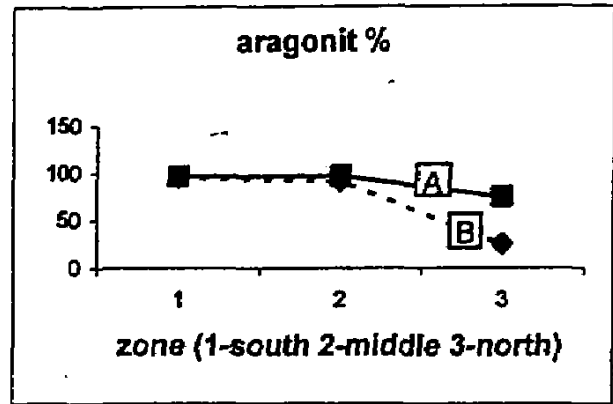
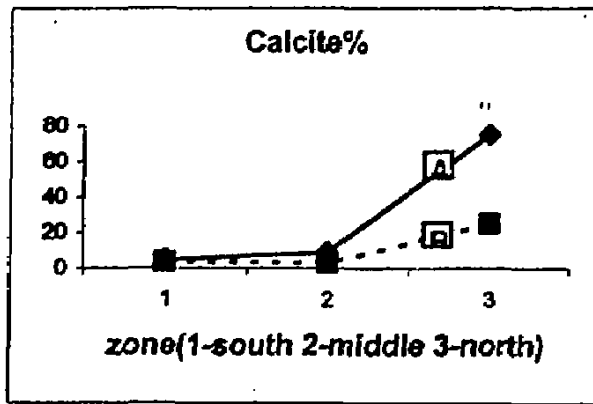


Fig. (1) : The Damietta Nile branch map showing sampling locations

Figure(2): The zonal distribution of environmental parameters of Damietta Nile branch molluscan shells and water(aragonite%, calcite%, chlorosity mg/l, alkalinity mg/l and pH value) A-bivalve B-gastropod



Figure(3): Zonal average distribution of trace metals(ppm $\times 10^{-3}$) in the studied shells of *Anodonta implicata* (bivalve A= adult, B=nepionic).

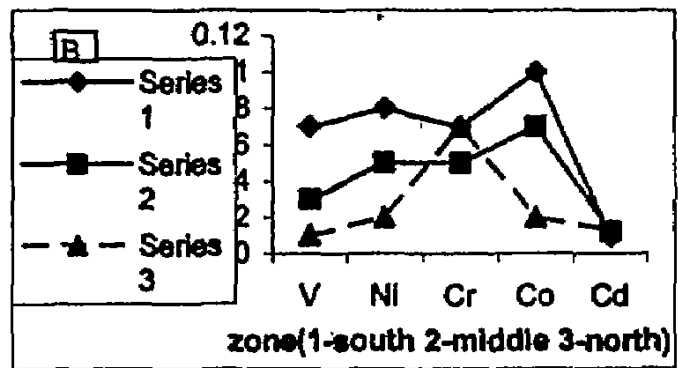
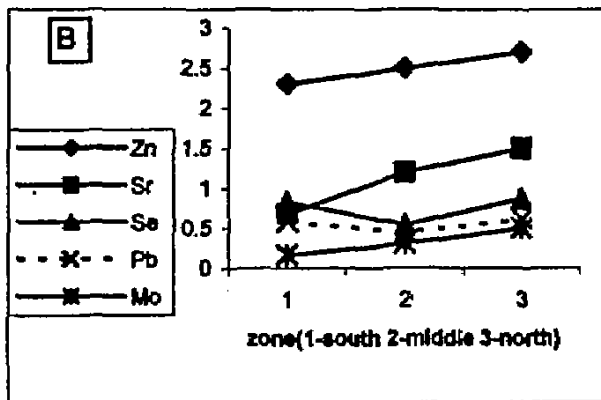
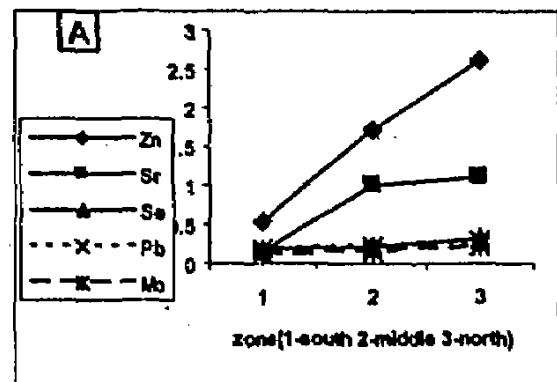
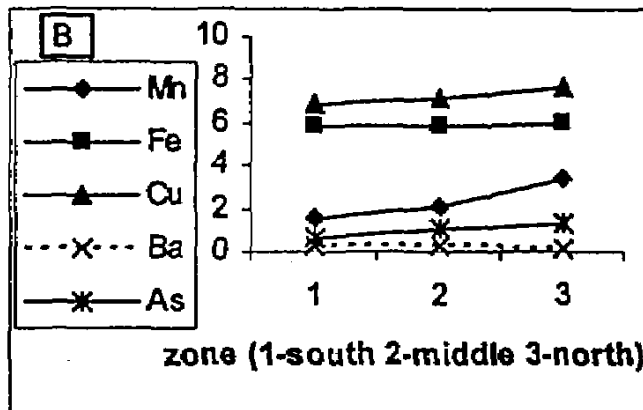
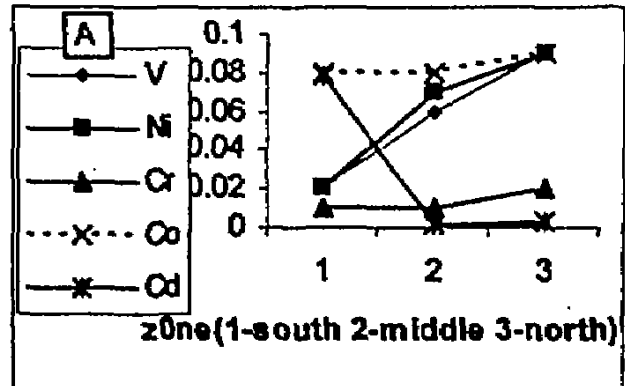
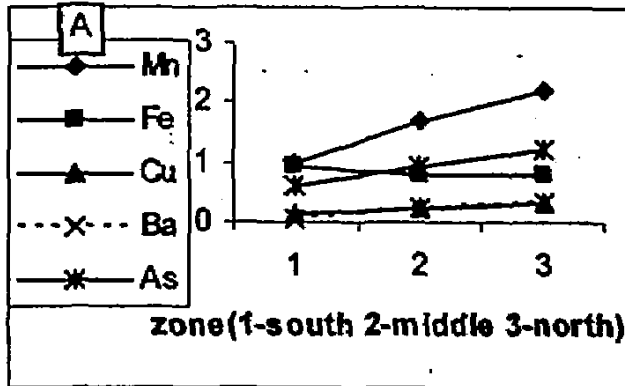
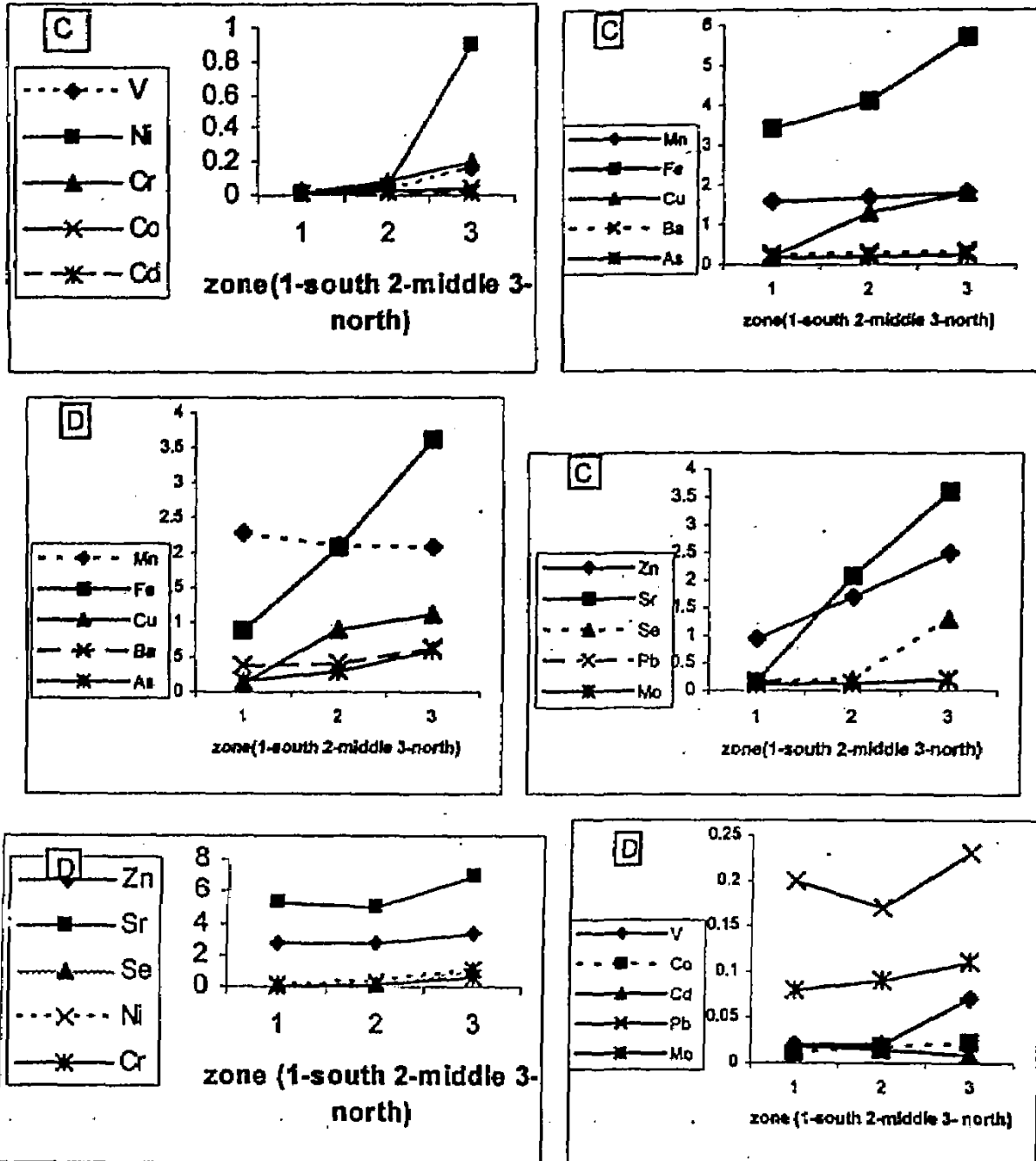


Figure (4): Zonal average distribution of trace metals(ppm $\times 10^{-3}$) in the studied shells of *Bellamiya unicolor* (gastropod A= adult, B=nepionic).



TRACE METALS IN MOLLUSCAN SHELLS FROM DAMIETTA NILE BRANCH SEDIMENTS

Figure(5): Zonal average distribution of trace metals(ppm and all values $\times 10^{-3}$) in the studied shells of bivalve and gastropod from lake Qarun.

