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DISTRIBUTION OF PHOSPHORUS FRACTIONS AND SOME OF HEAVY METALS IN SURFACE SEDIMENTS OF BURULLUS LAGOON AND ADJACENT MEDITERRANEAN SEA

KHALIL, M. KH.; RADWAN, A. M. AND EL-MOSELHY, KH. M.*

National Institute of Oceanography and Fisheries, Alexandria, Egypt. khalid19670@yahoo.com

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ABSTRACT

Twelve samples were collected from the surface sediments of lagoon and adjacent Mediterranean Sea. Samples were analyzed for grain size, total organic carbon (TOC), total carbonate (CaCO₃), various forms of phosphorus and heavy metals (Fe, Mn, Zn, Cu, Ni, Co, Cd and Pb). The results indicated that the sediments were composed of sand, silt and clay enriched with carbonate (1.95 – 34.1%) and total organic carbon (0.06 and 3.62%). The total-P content ranged between 58 and 1186 μ g/g, and the fraction associated with apatite minerals (P_{HCl}) was the dominant (it constitutes about 50 – 73% of TP) implying that it was the main storage of phosphate. The various P phases demonstrated significant positive correlation with mean size, silt, clay, TOC, CaCO₃ and TP, while only P_{ex} was correlated with the metals. As well as, the obtained data indicated that the variations of the measured metals in sediments are varied depending on the locations, whereas the high levels were observed in the western area of the lagoon. The studied gave positively correlated with each other and organic carbon suggested that the distributions of these metals are associated with the organic matter accumulation.

1. INTRODUCTION

Burullus lagoon is one of four shallow brackish water coastal lagoons (namely: Manzala, Burullus, Edku and Maryout) of Nile Delta. It is the second largest one which has an area of about 410 Km². It lies between longitudes 30° 30′ and 31° 10′ E and latitudes 31° 21′ and 31° 35′ N. `The lagoon width varies between 4 and 16 Km, and its mean water depth varies from 60 to 150 cm. It is separated from the Mediterranean Sea by sand bars and sand dunes of different widths and heights and is connected to the sea with Boughaz El-Burullus. Burullus lagoon is affected mainly by agricultural drainage water mixed with different types of wastes from fish farms (Terra drain, drain 7, drain 8 and drain 11), wastewaters effluents (Terra drain, drain 7 and El-Gharbia drain) as well as domestic drainage water discharged mainly from El-Gharbia drain and drain 11.

The largest pool of phosphorus within a lagoon is usually associated with sediments, which play a major role in the phosphorus Sediment bounds phosphorus constitutes a vast source of potential nutrient for phytoplankton, macrophytes, and benthic algae. The availability of phosphorus from this pool depends on the ability of sediments to retain or release phosphorus prevailing environmental conditions (Pettersson, 1986). Böstrom et al. (1982) reported that the aerobic sediments in ologotrophic lakes have been considered as a sink for phosphorus, while anaerobic sediments in very eutrophic lakes have been regarded as a source of nutrients. During decomposition of organic matter in oxidizing sediments, part of the released phosphate adsorbs on Feoxihydroxides, carbonates and clay minerals (de Lange, 1986; Lucotte and d'Anglejan, 1988). The forms of phosphorus in sediment proved useful understanding how sediment releases phosphorus into, or takes up phosphorus.

Sediments can act as a sink and a possible source of metals for the environment. Some of heavy metals are regarded as serious pollution of aquatic ecosystem because of their environmental persistence, toxicity and ability to be incorporated into food chains (Förstner and Wittman, 1983).

The present study investigates the geochemical and grain size characteristics of sediments in Burullus lagoon. It is based on determination the distribution of various forms of phosphorus and the concentrations of metals (Fe, Mn, Zn, Cu, Ni, Co, Cd and Pb), to asses their potential biological effects and the extent to which this distribution is related to other sediment components such as mean size, total carbonate and organic carbon.

2. MATERIALS AND METHODS

Surface sediment samples were collected during January 2006 from twelve stations, which covered all parts of Burullus Lagoon and nearby water of the Mediterranean Sea (Fig. 1).

The sampling stations were divided as follow: Station 1: represents lagoon-sea communication. Stations 2 and 4: represent the drains (7 and 8). Stations 3 and 5: represent southern part (middle sector) of the lagoon. Station 6: represents eastern sector of the lagoon. Station 7: represents northern part (middle sector) of the lagoon. Station 8: represents western sector. Stations 9 – 12: represent marine area of the Mediterranean Sea.

Grain size analysis was carried out by dry sieving for sand and by pipette technique for silt and clay fractions (Folk, 1974). Total organic carbon (TOC) measured by using dichromate wet oxidation method (Walkely and Black, 1934). Carbonate contents were

determined by titration technique (Black, 1965 and Berner: 1971,). Total metal concentrations were measurement were done using blend of acids HF-HCl-HNO₃ then the digested samples contain the metal were measured by using flame-atomic absorption spectrophotometer, Perkin Elmer, model Aanalyst 100. The samples were analyzed for different forms of phosphorus using an extraction procedure based mainly on the methods of Aspila et al. (1976) and de Lange (1992). In this procedure, the original sample was separated into two weighted sub samples. One of each was burned at 550 °C, and then its ash was extracted with 1 N HCl to determine the total phosphorus (TP). The second subsample was extracted with 2 N NH₄Cl solution, which represented the exchangeable and carbonate-associated phosphate (Pex). The residue from NH₄Cl extraction was extracted with 0.1 N NaOH solutions to give fraction of iron and aluminum-associated phosphate (P_{OH}). The residue from NaOH extraction was then extracted with 0.5 N HCl, which represented calcium-associated phosphate/apatite (P_{HCI}). The remaining sediment from the last extract was done in a way similar to that for the determination of TP, represented the residual phosphate (P_R).

3. RESULTS AND DISCUSSION

3.1. Grain size

The obtained data of grain size revealed that the sediments collected from the present investigated area composed of an admixture of sand silt and clay, having mean size varied between 1.26 Φ at station 10 (Mediterranean Sea) and 8.58 Φ at station 7 (Burullus Lagoon). The sand rich sediments prevailed in the lagoon-sea communication, which is derived mainly from the Mediterranean Sea and marine sediments (Table 1). Whereas, silty clay rich sediments prevailed in the eastern and southern parts of the lagoon,

which transported by the drains and canals. Moussa (1984) mentioned that the microscopic of sands in Lake Burullus showed that the sand fractions larger than 125 microns were entirely of mollusk shells and shell fragments, ostracods and foraminifers.

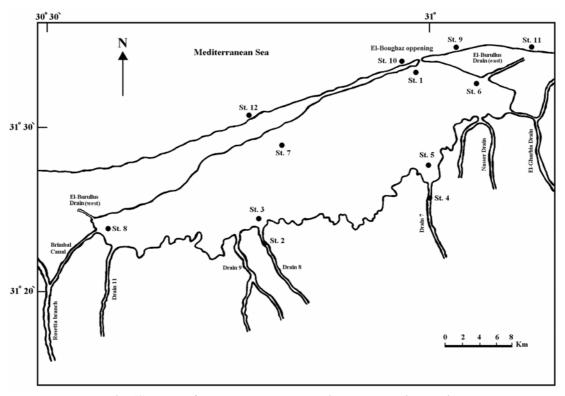


Fig. (1): Map of Burullus Lagoon showing the sampling stations.

Table (1): Grain size composition, total organic carbon (TOC) and carbonate (CaCO₃) of sediments collected from Burullus Lagoon and nearby Mediterranean Sea.

Stations	Mean size (Φ)	Sand %	Silt %	Clay %	Sediments type	TOC %	CaCO ₃ %
1	4.95	37	44	19	sandy silt	1.74	13.53
2	8.37	2	38	60	silty clay	1.75	6.59
3	7.7	7	46	47	silty clay	1.58	13.53
4	8.41	3	36	61	silty clay	1.13	7.68
5	6.99	20	35	45	silty clay	2.58	34.1
6	6.26	26	34	40	silty clay	1.01	18.94
7	8.58	7	20	73	clay	0.83	11.58
8	5.43	29	33	38	silty clay	3.62	24.14
9	2.46	100	0	0	sand	0.06	2.27
10	1.26	100	0	0	sand	0.15	3.78
11	1.72	100	0	0	sand	0.06	3.61
12	2.84	100	0	0	sand	0.18	1.95

3.2. Total organic carbon (TOC)

Content of TOC was highly variable in the collected sediments samples at the different sampling stations. In general, the coarser sediments of the marine samples had low TOC contents, which varied between 0.06 and 0.18 % (Table 1). While the finer sediments of the lagoon and drains were significantly enriched with organic carbon. The highest value (3.62 %) was recorded at st. 8 (western sector), reflecting high rate of organic matter accumulation from the domestic drainage water. According to Muller and Suess (1979), the ratio between the recycled and accumulated organic matter in sediments is controlled by the bulk sedimentation addition, rate.In contribution of plant detritus from the nearby vegetable area. At station 7 (middle sector), the TOC concentration was partially low, due to the abundance of carbonate and its faraway from the direct effect of discharges originated from the southern margin of the lagoon. TOC showed positive correlation with both mean size of the particles and silt, in contrast it gave a significant negative correlation with sand percentage (Table 2). These results were agree well with the relationships reported by Moussa et al. (1994) for Lake Edku sediments with mean size varies from medium sand to silty clay. Where the silt fraction could be clay firmly cemented by organic matter or that the organic detritus predominates in the silt size range.

3.3. Carbonate

In general, the carbonate content in sediments of Burullus lagoon and nearby Mediterranean Sea varied between 1.95 and 34.1% (Table 1). The lowest values of carbonate (1.95-3.78%) were found in the marine sediments where quartz sand was predominates. As well as, carbonate content

in sediments from the drains showed values (6.59 and 7.68 %) lower than those recorded inside the lagoon. It is thus evident that the shells of organisms are the principle contributors of carbonate in sediments. A direct significant correlation (r = 0.61) with the silt fraction (Table 2) may be attributed to the presence of carbonate debris in silt size.

3.4. Phosphorus Forms

Levels of total and different forms of the phosphorus in the sediments collected from Burullus Lagoon and its nearby waters of the Mediterranean Sea are given in Table (3).

3.4.1. Total phosphorus (TP)

The concentration of total phosphorus varied between 58 and 1186 µg/g. The lowest phosphorus content were generally characterized the marine sediments, due to the notably higher amounts of sand and lower of organic carbon. concentrations were found in the vicinity of lagoon-sea communication area (st. 1) mainly due to the predominance of quartz sand at this area which resulted in low phosphorus concentrations. However, the substrate of st. 7 is muddy of moderate level of TP (421 µg/g) due to its faraway from the sources of drainage waters and big aquatic plant area at this station. McRoy and Barsdate (1970) and McRoy et al. (1972) assumed that aquatic plants are very effective in taking up sediment phosphorus. Moreover, relatively lower organic matter in this area may deprive the sediments of phosphorus (Balzer et al., 1986). The high phosphorus content $(967.0 - 1014 \mu g/g)$ found in the southern area of the lagoon directly affected by drainage water. In this context, the sediment samples collected from the drains recorded the highest values of TP (1128 -1186 $\mu g/g$).

	M. size	Sand	Silt	Clay	TOC	CaCO ₃	TP	Pa	Рон	Рна	PR	Pmm	PO	Pb	Cu	Zu	Fe	Mn	z	8
M. size	1.000																			
Sand	-0.973*	1.000																		
Silt	0.803*	+968.0-	1.000																	
Clay	0.974	-0.952*	0.717*	1.000																
TOC	0.525	-0.647*	0.735*	0.508	1.000															
CaCO,	0.448	-0.547	*809.0	0.439	0.810*	1.000								*						
T	0.854*	+098.0-	0.847*	0.766*	0.590	0.474	1.000													
D	0.416	-0.551	0.686*	0.392	0.825*	0.556	0.440	1.000												
Post	0.827*	-0.805*	0.711*	0.774*	0.380	0.142	0.903*	0.264	1.000											
Puc	0.851*	-0.898*	0.962*	0.747*	0.703*	0.633*	0.928*	0.594*	0.761*	1.000										
Po	0.846*	-0.871*	0.891*	0.753*	0.604*	*109.0	0.961*	0.428	0.810*	0.965*	1.000									
P.	0.882*	-0.913*	0.935*	0.789	0.650*	0.548	0.973*	0.524	*098.0	0.984	0.981	1.000								
Cd	0.055	-0.167	0.211	0.117	0.713*	0.427	0.035	*849.0	-0.010	0.117	-0.007	0.081	1.000							
Pb	0.050	-0.162	0.201	0.115	0.705*	0.427	0.031	0.664*	-0.011	0.111	-0.006	0.077	*6660	1.000						
Cu	0.110	-0.216	0.249	0.168	0.735*	0.434	0.087	0.692*	0.043	0.164	0.037	0.130	0.995*	0.993*	1.000					
Zn	0.080	-0.190	0.230	0.139	0.723*	0.446	0.054	0.688*	-0.001	0.141	0.011	0.101	0.997*	.966.0	.866.0	1.000	-			
Fe	0.002	-0.116	0.295	-0.022	0.273	0.237	-0.072	0.613*	-0.258	0.192	0.049	0.075	0.155	0.151	0.149	0.155	1.000			
Mn	0.012	-0.126	0.176	0.077	*869.0	0.411	0.004	0.663*	-0.043	0.083	-0.036	0.047	*866.0	*866'0	*066.0	0.992*	0.172	1.000	100000	
ž	0.079	-0.189	0.228	0.139	0.718*	0.441	0.046	*169.0	-0.010	0.137	0.005	0.095	0.997*	0.995*	*266.0	1.000*	0.177	0.992*	1.000	-
Co	. 0.072	-0.182	0.221	0.134	*902.0	0.412	0.037	*1690	-0.003	0.124	-0.010	0.086	*866.0	*966.0	*966.0	*8660	0.173	0.993*	.0.999*	-

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5/TP % Table (3): Level of phosphorus forms (μg/g) in sediment samples from Burullus Lagoon and nearby Mediterranean Sea 4/5 % 29 29 22 22 20 20 3/5 % % 2/5 1/5 % P_{sum} (5) 224.8 P_R (4) P_{HCI} (3) Poh (2) Pex (1) TP Stations

3.4.2. Fractionation of phosphorus

Par

The values of P_{ex} ranged from 2 µg/g (st. 10) to 36 μ g/g (st. 8). The distribution pattern of Pex for the surface sediments was quite similar to that of silt and TOC, the respective correlation coefficients were significant (r = 0.69 and 0.82) respectively. Also, significant positive correlations were found with all studied metals (Table 2). The high value of P_{ex} at station 8 was attributed mainly to the domestic drainage and wastewaters from drain 11. Pettersson (1986) showed that sediments characterized as gyttja (high TOC) or clayish gyttja in lakes that were former recipients of municipal sewage had a large proportion of loosely sorbed P which is easily released to the lake water. In laboratory experiments, the clayish gyttja sediments released P under all simulated conditions (aerobic/ anaerobic, pH 8.0 to 10) (Böstrom, 1984).

PoH

P_{OH} concentrations ranged from low value of 7 µg/g at station 10 to a high value of 194 µg/g for sediments of station 2. In general, the low concentrations were found in the lagoon-sea communication area and marine sediments. Whereas, the high levels were recorded at stations 2, 3 and 4 (southern part of the lagoon and its drains). Table (3) show that P_{OH} is related to the sum of different P fractions, but is not related to total Fe content of sediment. The lack of correlation with Fe caused by the large amount of Fe to which no phosphate is likely to be associated, such as magnetite-Fe (reported for the volcanic turbidites) and pyrite-Fe (reported for the organic-rich turbidites) (Wensink, 1984; De Lange et al., 1987). The P_{OH} was insignificantly related with CaCO₃ and with each of Fe and Mn (r = 0.14, -0.26 and -0.04, respectively). El-Rayis et al. (2005) reported similar pattern for Abu Oir Bay with possible release of considerable portion of these metals from the sediments after conversion to the more soluble reduce Fe (II) and Mn (II) from sediments. So the association of PO₄ in this fraction (P_{OH}) is expected to be more due to Al rather than with Fe (Chen *et al.*, 1973).

PHCI

P_{HCl} concentrations ranged from low value of 44 µg/g at station 10 to a high value of 443 µg/g at station 3. It represented about 50 - 73 % of the P_{sum} . Therefore, it is the main storage of the total phosphorus in the sediments. De Lange (1992); Khalil (2003) and El-Rayis et al. (2005) stated that the apatite-P content exhibited the majority of all extracted P forms in Madeirs Abyssal Plain, Manzala Lagoon and its neighboring marginal Mediterranean Sea and Abu Qir sediments, respectively. Apatite-P showed significant positive correlation with TP content (r = 0.93). As well as, the silt content was shown to play a principal role in determining the concentration levels of apatite-P (r = 0.96) (Table 2).

P_R

Residual phosphate was the second abundant after P_{HCl} , it represented about 12-29 % of the P_{sum} . Its concentrations ranged from 7 $\mu g/g$ (station 10) to 209 $\mu g/g$ (station 2).

Fig (2) shows the regression line between total phosphorus and the sum of extracted P (P_{sum}). The relationship showed significant positive correlation (r = 0.97, slope = 0.7) representing good agreement between both values (P_{sum} and TP) along the all area of the present study. As well as, mean value of P_{sum} exhibited a good recovery (85 %) in relation to the TP values. In this context, Penn *et al.* (1995) and El-Rayis *et al.* (2005) gave a recovery of 76.3 % and 85 %, respectively.

In general, most of the various P phases demonstrated significant positive correlation with mean size, silt, clay, TOC, $CaCO_3$ and TP. In contrast, it showed significant negative correlation with the sand percentage. While, relationships with metals showed insignificant correlation except for P_{ex} which gave significant positive correlation with all studied metals.

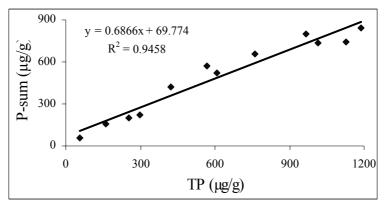


Fig. (2): Regression line of the relationship between sum of extracted phosphorus and total-P

3.5. Heavy metals

3.5.1. Fe and Mn

The Fe concentrations are nearly the same for different stations (Table 4), and ranged from 2551 to 3816 $\mu g/g$. The Mn content is the highest (748 $\mu g/g$) at station 8, while the whole studied stations showed slight variation, which ranged from 61.76 to 85.14 $\mu g/g$.

Except the western sector (st. 8), Fe/Mn ratio in the sediment samples was about 43.5 for other parts of the lagoon and marine area suggesting a dominating manganese source from terrestrial one (from lithogenous phase). At st. 8, the Fe/Mn ratio was 4.2; may be suggesting a dominating manganese source other than the terrestrial one, which resulted from chemical precipitation of Mn from water or due to the high Mn content at this station.

3.5.2. Zn and Cu

The present study showed moderate concentrations of Zn and Cu in the sediments

from the eastern and middle areas of Burullus lagoon. It ranged from 28.40 to 77.26 µg/g for Zn and 25.45 to 41.47 µg/g for Cu (Table 4). On the other hand, higher concentrations of 754.59 and 366.35 μ g/g for Zn and Cu respectively were reported in the sediments of the western area (st. 8). The lower concentrations were recorded for stations 10. 11 and 12 (Mediterranean Sea). This agrees with Radwan and Shakweer (2004), where the levels of trace metals in the bottom sediments of Lake Burullus decrease from west to northeast. Both Zn and Cu were probably associated with the hydrogenous manganese phase, where significant positive correlation was detected between them and Mn, Pb and Cd. In the meantime, Zn and Cu were also positively correlated with TOC, Ni and Co. Lynn and Bonatti (1965) suggested increased copper and zinc the concentrations in the oxic sediments were a consequence of manganese oxide dissolution in the subsurface resulting in their liberation and subsequent precipitation in the surface layer. Hutchinson (1975) reported that water rich in brown humic matter in content with mud particles of lake water takes up Cu more than water poor in s uch matter.

Table (4): Level of heavy metals (μg/g) in sediment samples from Burullus Lagoon and nearby Mediterranean Sea.

Stations	Fe	Mn	Zn	Cu	Ni	Co	Cd	Pb
1	3816	85.14	67.69	32.41	34.46	7.82	2.15	10.34
2	2942	70.28	28.40	35.66	12.95	3.45	1.36	9.01
3	2551	61.76	80.71	41.47	32.43	7.50	2.55	13.66
4	2737	64.11	77.26	39.67	34.54	8.41	2.13	11.46
5	2909	72.68	68.14	32.01	27.95	4.38	1.88	10.53
6	3115	77.74	54.42	25.45	25.81	4.79	1.81	15.65
7	3054	65.77	70.52	36.93	34.65	7.67	2.06	12.12
8	3106	748.00	754.59	366.35	309.56	68.34	24.68	153.07
9	2816	66.27	75.26	42.04	32.74	5.99	1.50	9.28
10	2685	56.27	7.89	2.67	2.92	1.42	0.75	4.51
11	3072	63.16	21.63	7.48	14.75	4.22	1.40	8.51
12	2742	64.69	11.29	3.39	7.68	2.44	1.14	7.00

3.5.3. Ni and Co

The present data depicted that the sediments content of Ni and Co attained its minimum values for the marines (Table 4). High values were observed at stations of the eastern and middle areas, while the highest extreme concentrations were for the western area (309.56 and 68.34 µg/g) for Ni and Co respectively. Ni and Co were closely correlated with TOC and Mn contents (Table 2). Nickolls and Loring (1962) reported correlation between Ni and organic matter in the British sediments. El-Ghobary and Latouch (1982) reported that large fraction of Ni may be removed from the sea water by scavenging action of Fe and Mn oxides (hydroxides). Francois (1988) showed that Ni distribution was primarily controlled by the mineralogical of sediments more than its association with organic matter accumulation. Surveying the correlation (Table 2), pointed that Ni and Co were also closely correlated with Zn, Cu, Cd and Pb.

3.5.4. Cd and Pb

Both showed the same pattern of distribution as recorded for the other studied metals. The high values (24.68 and 153.07

µg/g) for Cd and Pb respectively were recorded at the western area. While, the lower values represented the marine area (Table 4).

In general, the present study showed that the high concentration of Mn, Zn, Cu, Ni, Co, Cd and Pb were recorded at the western area of the lagoon; due to the fresh water discharged from Brimbal canal, beside the domestic and wastewater discharged from drain 11. Also, this area covered by *Eichhornia crassipes* (floating plant) which contain high levels of metals (Shakweer and Radwan, 2004).

Possible Biological Effects

Heavy metals are regard as serious pollution of aquatic ecosystem because of their environmental persistence, toxicity effects on living organisms. To estimate the biological effects of metals, ERL (Effects-Range Low) and ERM (Effects-Range Median) reported by Long and Morgan (1990) and Long *et al.* (1995) were used (Table 5). It can be observed that the ERL and ERM delineated concentrations at which adverse biological effects rarely occur (< ERL: about 56.7% occurrence), occasionally (between ERL and ERM: 35% occurrence), and frequently(>ER about 8.3% occurrence).

Table (5): Number of samples that had metal concentrations above the sediment effects data of ERL and ERM in Burullus Lagoon and nearby Mediterranean Sea.

	Cd	Pb	Cu	Zn	Ni
Below ERL	2	11	6	11	4
Between ERL and ERM	9	0	5	0	7
Above ERM	1	1	1	1	1
ERL	1.2	46.7	34	150	20.9
ERM	9.6	218	270	410	51.6

The concentrations of Cd and Ni in the most sediment samples were between ERL and ERM, which indicated possible detrimental effects to benthic organisms. While, the concentrations of Pb, Cu and Zn in the most sediment samples were below the ERL suggesting that the sediment samples may not have adverse effects on the benthic organisms. The western basin of the lagoon had Cd, Pb, Cu, Zn and Ni concentrations above the ERM of Long and Morgan (1990) and Long *et al.* (1995) suggesting that is deemed detrimental to benthic organisms.

4. SUMMARY AND CONCLUSION

The present investigation indicated that the sediment samples from the Mediterranean Sea showed the lowest values of TOC, carbonate and P forms, while the western sector of Burullus Lagoon exhibited the highest ones. The various P phases recorded significant positive correlation with mean size, silt, clay, TOC, CaCO₃ and TP, and $P_{\rm ex}$ only showed significant positive correlation with metals. The apatite ($P_{\rm HCl}$) is the main storage of P and is the most stable form of P-mineral under oxic and anoxic environmental conditions. The order of abundance of the P fractions in Burullus lagoon sediments were as follows $P_{\rm HCl} > P_{\rm R} > P_{\rm OH} > P_{\rm ex}$.

As other studied parameters, high concentrations of Mn, Zn, Cu, Ni, Co, Cd and Pb were recorded at the western area of the lagoon, which attributed to the fresh water discharged from Brimbal canal, beside the domestic and wastewaters discharged from drain 11. Also this area covered by

Eichhornia crassipes which contain high levels of metals. Except Fe, the studied metals gave significant positive correlation with each other, and in the meantime were positively correlated with organic carbon suggested that the distribution of these metals are association with the organic matter accumulation.

Most of the sediment samples from Burullus lagoon appear to have heavy metal concentrations below the threshold that were believed to be safe to benthic organisms.

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