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Environmental Geochemistry and Fractionation of Cadmium Metal of Brullus Lagoon, Egypt

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Abstract : The pollution of water and surficial bottom sediment of the Brullus lagoon is indicative of both water and food web quality in general. Fourteen samples were collected from surface water sample and bottom sediments sample among sites covering the Brullus Lagoon during summer 2014, in addition to seven bottom sediment sample collected from the sits near outlet of agricultural drains. A few research were carried related to study of fractionation of Cadmium elements, so, the main objective of this study is assessment of environmental for this toxic element through the Egyptian standards and the world-wide organizations. Analytical techniques have been utilized to analyze cadmium content. The sequential extraction of the Cadmium Metal is conducted on representative samples in order to assess the potential mobility and bioavailability of this metal in the studied sediments. The present study documents hard pollution by Cd, possibly as a result of using fertilizer. The territory around inlet and southeastern drain show obvious pollution by the studied metal. The main reason for such pollution resulting from industrial activities and agricultural drains. The disregards of the anthropogenic activities are the main reason of pollution in the studied lagoon. Among all the fractions for Cd, the reducible fraction was the most abundant pool, followed by exchangeable, carbonate, organic and finally Fe- Mn hydroxide fraction. Calculated pollution indicators encompass contamination factor and geoaccumulation index were reflects high level of pollutants in the vicinity of the agricultural activity at the northern areas, also, adjacent to southern drains. Oversight permanent and Bureaucracy program for monitoring the abundance and distribution of toxic metals in the studied lake should be enjoined.
Keywords : Brullus lagoon, bottom sediments, pollution, cadmium content, Fractionation of Cadmium, Egypt.

Significance Statement:

- This paper is interested with the study of the distribution of cadmium in water and bottom sediments of Brullus Lagoon
- The average content of Cd in the studied water (0.9 ppm) andin sediments (24.85ppm)
- The relative order of abundance of cadmium fractionation in bottom lagoon sediments is reducible > exchangeable > carbonate > organic > and finally Fe- Mn hydroxide fraction
- The highest values for pollution due to agricultural and industrial drains

Introduction

Brullus lagoon is forming a part of a Nile delta system where considered as one of coastal shallower lakes at the northern Egypt. It is located between longitudes 30°31` and 31°05` E and latitudes 31°25` and 31°35`. It occupies an area of 420 km², has length is about 53 km, and its width is about 13 km. It connects to the sea through Boughaz El- Brullus at its northeasternportion.

The drains are the main feeder of the lagoon, where it is consider as a reservoir for the collection of sewage water (agricultural, fish farms and industrial drainage). The lagoon receives drainage water from southern boundary through seven drains, drain 7,8,9 and 11 in the southern side, Terra drain and El-Gharbia drain at the south eastern side of the lake , It also receives fresh water Through the Brimbal Canal located in the far west of the lagoon and represent the waters of the mouth of the Rashid branch .

The environment of Brullus Lake has changed during the last three decades as a result for constructing several drains to convey agricultural wastes into the lagoon. In addition, large area has been dried up to agricultural lands encompass the southern and eastern fringes of the lake.

Cadmium can mainly be found in the earth's crust. A great amount of cadmium is released into the environment, rivers through weathering of rocks and the ground because it is found in manures and pesticides. Cadmium waste streams may also enter the air through human activities, such as manufacturing, waste combustion and burning of fossil fuels. Human exposure to cadmium fundamentally through food is causing bioaccumulation in bodies. Zielhuis[1], gave examples for organs which influence by the bioaccumulation of toxic concentrations of cadmium Table 1.

Table1. Examples for organs which influence by the bioaccumulation of toxic concentrations of cadmium .Zielhuis[1]

System - Organ	General Health Effects
Renal System	Tubular, glomerular damage, proteinuria
Blood System	Slight anemia
Respiratory Tract	Emphysema
Prostate gland, lung	Cancer
Skelton	Osteomalacia (itai-itai)
Chromosomes	Aberration

Cadmium metal can be emitted into a lagoon environment by human activities. They have been introduced into ecosystem from industrial and agricultural operations. Some uses studied through which Cadmium metal enter to aquatic environment as a result of indusial processes. examples of the various uses of cadmium are Alloys, Ni / Cd Batteries, coal combustion, chemicals, pharmaceuticals, dental, coatings (anticorrosive), fertilizers, fossil fuel combustions, paint and pigments, plasticsSiegel[2].

The ecology of Burullus Lagoon has been studied by many numerous publications, e.g., Dewidar and Khedr[3], Al-Sayes[4]*et al.*,Chen5, et al., Mohamed[6]**,** *et al.*,Mamdouh[7]**,** *et al.*, El-Asmar[8], *et al.*,Khalil and El-Gharabawy[9], among others. Many authors study metal speciation in aquatic ecosystems (Vicente et al[10].Nsikak, et al[11].,Essien et al.[12], Ramadan[13], Prusty et al.[14], Weng, et al.[15], Jain, *et al.*[16],Zakir[17],Zakir, *et al.*[18],andLasheen, *et al*[19]).

The environmental problems of the lake originated mainly from the man-made influences; on the other hand, the lake seems to be distinct in terms of waste material receives. The present study is throwing more light to map water and bottom sediment pollution in Brullus lagoon using chemical analysis.

Materials and Methods

Fourteen sites covering the Brullus Lagoon body were specified during summer 2014, and each site was represented by a surface water sample and a bottom sediment sample, in addition to seven bottom sediment samples collected from the sits near outlet of agricultural drains (Figure1). The preparation of sediment for trace metal determination was accomplished according the procedure described in FAO technical paper No.158.

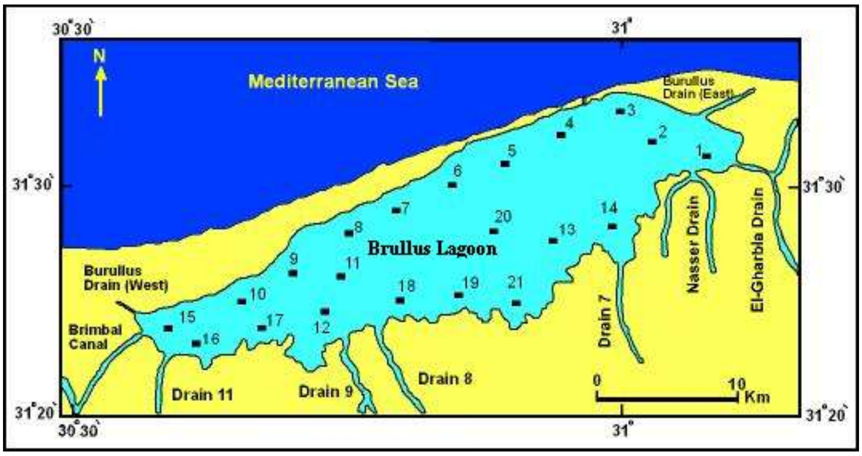

Figure (1): Location map of the sampling sites.

Sediment samples were air dried, disintegrated and representative quotient was totally grind, then preserved for chemical analyses. The chemical analyses were preceded on whole sediment samples for Cd using atomic absorption spectrometry (Perkin-Elmer 3110, USA) with graphite atomizer HGA-600, after using the digestion technique according to the standard APHA[20].

Sequential Extraction Procedure:

The sequential extraction of the Cadmium Metal is conducted on representative samples in order to assess the potential mobility and bioavailability of this metal in the studied sediments. The sequential extraction scheme according to Tessier[21].One gram of each sample is weighed and the following five steps are obtained:

- Step 1. The exchangeable fraction is extracted with 8 ml of 1M 2MgCl at neutral pH for 1 h.
- Step 2. The carbonate-bound fraction is extracted with 8 ml of 1 M Sodium acetate adjusted to pH 5.0 with Acetic acid (for 5 h).
- Step 3.The Fe-Mnoxyhydroxides- bound fraction is extracted with 0.04 M Hydroxylamine hydrochloride in 25% Acetic acid (v/v) at 96°C with occasional stirring for 6 h.
- Step 4.The organic-bound fraction is extracted with 3 ml of 30% Hydrogen peroxide in 0.02 M Nitric acid (adjusted to pH 2 with HNO). The mixture is heated to 85°C for 2 h with occasional stirring.
- Step 5. The Residual / lithogenic fraction is obtained by complete digestion of the residue with a mixture of 3 HF-HCI/HNO in a digestion bomb. The overall recovery rates of the analyzed heavy metals range from 90 to 110 %.

Cadmium content in water lagoon.

The cadmium content of the investigated water is given in (Table 2).Cadmium concentration ranges from 0.2 to 3.2 µg/L, with an average 0.9µg/L. (Figure 2).

Table 2: Concentrations of Cadmium in waterµ g/L, bottom sediments ppm and five steps of Cd fractionations data of bottom Brullus Lagoon sediments.

	Total Cadmium content		**Cadmium fractionations in sediments ppm**				
Stations	In water µ g/l	In sediments ppm	(EXC) Exchangeable	(CA) Carbonate bond	(FM) Fe-Mn hydroxide bond	(OM) Organic bond	(RES) Insoluble
1	Nd	29	1.6	1.9	0.9	1.1	23.5
2	2.6	30	1.9	1.3	1.4	1.1	24.3
3	3.2	27	1.7	1.7	0.7	0.7	22.2
4	2.1	31	2	2.5	0.7	1.7	24.1
5	0.6	26	1.7	1.5	1.1	1.3	20.4
6	0.4	29	2.2	1.4	0.6	1	23.8
7	0.3	31	3.5	1.9	0.9	1.5	23.2
8	0.2	13	3.1	1	0.6	1.2	7.1
9	0.3	11	3	0.7	0.6	1	5.7
10	0.3	22	2.8	1.8	0.5	0.9	16
11	0.4	21	3	0.6	0.8	1	15.6
12	0.2	30	3.2	1.3	1.9	1.7	21.9
13	0.6	28	3	1.2	1.1	1.7	21
14	0.9	29	3.4	1.4	0.8	1.2	22.2
15	N.m	25	3.3	0.7	1.8	1.2	18
16	N.m	19	2.7	1.1	1.1	0.8	13.3
17	N.m	20	3.2	1.1	1.1	1.2	13.4
18	N.m	23	3	1.7	0.7	1.4	16.2
19	N.m	28	3.3	1.7	0.9	1.1	21
20	N.m	26	3.3	1.3	1	1	19.4
21	N.m	28	3	1.2	1.1	1.2	21.5
Average	0.9	24.85	2.82	1.36	0.97	1.2	18.52
Max	3.2	31	3.5	2.5	1.9	1.7	24.3
Min	0.2	11	1.6	0.6	0.5	0.7	5.7

Nd : under detected limit N.m: Not collected so, not measured

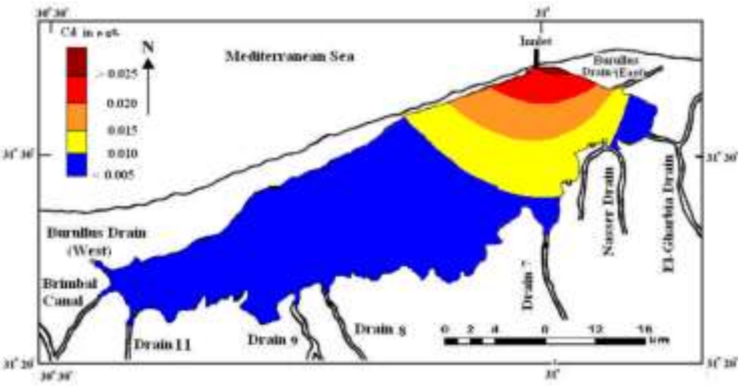

Figure (2): spatial distribution map of Cd value (µg/L) in the studied lagoon water

Along the Spatial direction of the lagoon, Cadmium contents increase toward northeastern directions. The data listed in table 2 indicate that the concentrations of Cd in lagoon water have concentrations higher than those in Mediterranean Sea water 0.00011 µg/L, which indicate the influence of the evaporation on the lagoon water. Generally the metal concentrations increase toward the northeastern area of the lagoon, possibly due to flocculation processes close to her inlet, addition to fossil Fuel combustions resulting from fishing boat and navigation process.

Geochemical backgrounds

The average earth's crust as quoted by McLennan and Taylor[22], comparison with the present data is given in Table 3. The rapprochement suggests the average content of Cd is more than 48fold the average earth's crust.

Table 3: Average Cadmium of the present work compared with average shale of Mason and Moor[23], Fresh water sediments (USPH)[24], McLennan and Taylor[22], Radwan[25, 26], McLennan and Murray [27], Mamdouh[28]. Masoud et al. [29] . All data are given in ppm.

Reference	Cd	Reference	Cd
Average shale of Mason and Moor[23]	0.2	Shale McLennan and Murray [27].	3
Fresh water sediments (USPH) [24]	1	Mamdouh[28]	2
Earth crust McLennan and Taylor[22]	0.5	Masoud et al. [29].	10
Radwan et al.[25]	5	Present study	25
Radwan[26]	7		

Cadmium metal in lake sediments The following is a short discussion on the environmental condition of cadmium content that may causes several hazard or vulnerable on human environment beside the geochemical distribution map. The Cd content of the investigated sediments is listed in Table 2.

Cadmium is considered as the most dangerous elements on aquatic life and humans. Cd exposure can cause both non carcinogen and carcinogenic risks such as kidney disease, skeletal damage, and even cancers Johri[30] et al. it is one of the black listed elements. The concentrations of cadmium in the bottom sediments of Brulluslagoon are given in Table 2. It ranged from 11 ppm to 31ppm, giving a general average of 24 ppm. According to Kabata-Pendias[32], the maximum permissible limit (MPL) of Cd in cultivated soil is 0.5ppm. The Department of Health and Human Services (DHHS) has determined cadmium and cadmium compounds as carcinogens; therefore, the Environmental Protection Agency (EPA) [31], has set a limit of 5 ppb of cadmium for drinking water. The average content of Cd in the study area is more than 48 fold the MPL of soil as quoted by Kabata-Pendias[32]. While the highest Cd content (31 ppm) is 62 fold. The frequency distribution of the Cd is unimodal with maximum at about 25-30 ppm (Figure 3).

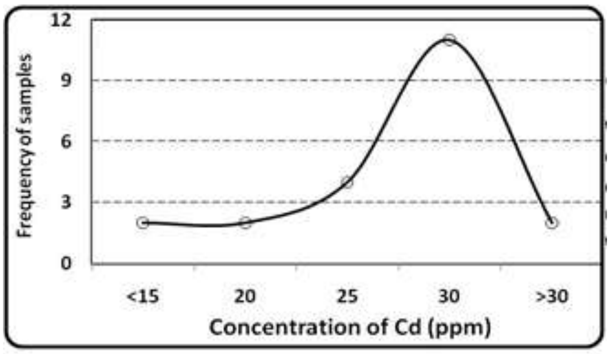

Figure 3: Frequency distribution of Cadmium in the studied lagoon.

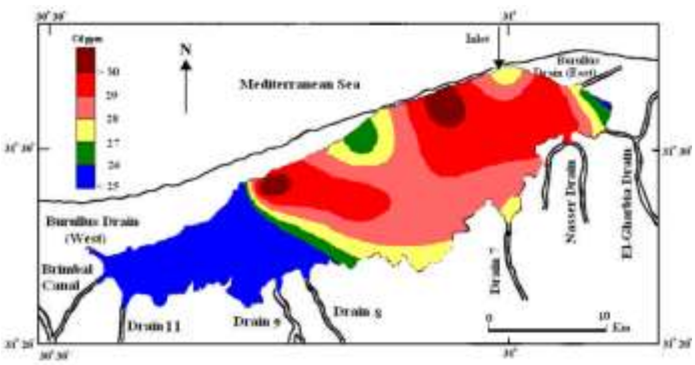

Figure (4): Geochemical map of Cadmium (in ppm), in the studied lagoon sediment.

Frequency and distribution data for cadmium content ppm for 21 bottom lagoon sediments are listed in Table 4.

It can be pointed out that, the most polluted sediments with cadmium giving high two concentration centers, located at the north area of the lake, where agricultural activity and the human activities at El-Maksaba and Mastarouh villages and toward the southeastern portion where water discharged from El-Gharbia drain. The distribution of cadmium is represented in Figure 4.

Table 4: Frequency & distribution data for Cd ppm for 21 bottom lagoon sediments.

Bin	Lower	Upper	Count	Percent	total	Percent
1	11	15	2	9.5	2	9.5
2	15	20	2	9.5	4	19
3	20	25	4	19.0	8	38
4	25	30	11	52.4	19	90.5
5	30	31	2	9.5	21	100

Table 5.Classification of risk assessment code (RAC).

Risk assessment code (RAC)	The sum of exchangeable and carbonate bound fractions
No risk	< 1
Low risk	1 - 10
Medium risk	11 - 30
High risk	31 - 50
Very high risk	> 50

Sequential Extraction of the Cadmium Metal:

Cadmium is found in various chemical forms in sediments and they show variable behavior in terms of mobility, bioavailability and toxicity Li. et al[33]. Based on their relative mobility in different chemical forms, bioavailability of heavy metals increases in the order: residual < organic < Fe- Mn oxy-hydroxides < Carbonate <exchangeable. Metals bound to the exchangeable fraction are easily available, whereas become more mobile and readily available with increasing acidity in the carbonate/adsorbed phases. The residual fraction having a little tendency to react chemically phase. The following is an attempt to throw more light on the manner by which the Cadmium metal is connected with the mineral constituents whether primary or weathering products.

Cadmium shows a distinctive partitioning pattern in the studied sediments. The residual fraction consider as the master portion and has accounts of the total Cd content approximately 74.5% (5.7-24.3 with an average 18.52 ppm), whereas exchangeable (1.6-3.5 with an average 2.82 ppm) , Organic bond (0.7-1.7 with an average 1.19 ppm) fractions, followed by the carbonate and Fe-Mnhydrooxide bond fractions. The results of cadmium metal partitioning in the examined sediments displayed in figure 5.

The residual and exchangeable fractions are the main modes of Cd content, while the average of bioavailable cadmium contents and the reducible fraction (comprised organic fraction and Fe- Mn hydroxide fraction) together represent about 25.5 % of total cadmium content and they could be related to anthropogenic activity. This conclusion agrees with Lashin and Ammar[34]. The association of Cd with the exchangeable phase agrees with Siegel [35] who noted that >20% of Cd is an available from easily soluble and exchangeable phases. According to Tessier[21] classification the acid soluble fraction includes the water soluble and exchangeable fractions. The cadmium fraction's is Precipitated or co-precipitated with carbonates by adsorption with weak electrostatic interactions on solid surfaces. This fraction constitutes the most mobile and potentially the most available cadmium species. The sum of exchangeable and carbonate bond fractions ranges between 3.2 to 5.4 ppm, with an average 4.1 ppm.

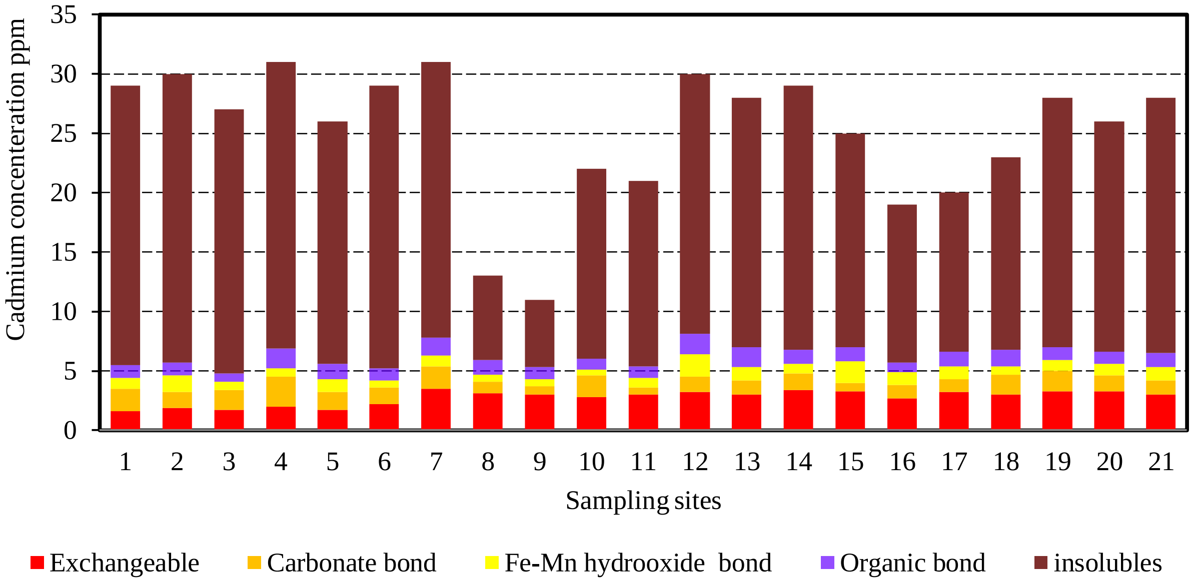

Figure 5: Comparison of the distribution of cadmium in various chemical fractions in the studied lagoon

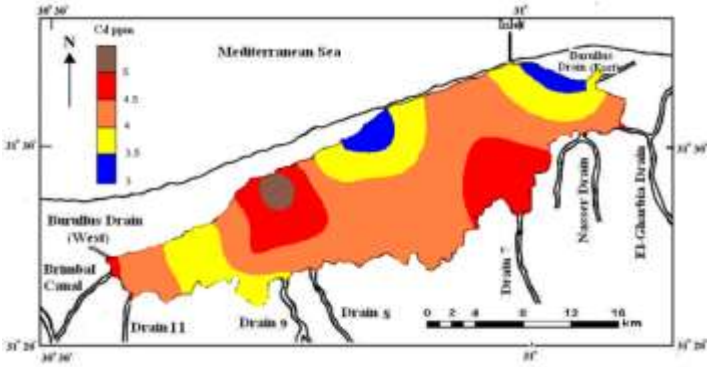

Figure (6): Geochemical mapof bioavailable content of cadmium (Sumexchangeable and carbonate fractions) in the studied lagoon sediment.

The most polluted lagoon sediments with Bioavailable content of cadmium giving high concentration at several sites, located at the north area of the lake, where agricultural activity at El-Maksaba and Mastarouh villages and toward the southeastern portion where water discharged from drain 7 and El-Gharbia drain. Also, adjacent to western drains (drain11 and brullus west drain beside Brimbal canal). Generally, distribution of cadmium bioavailable contents increased where agricultural drain which loaded by fertilizer, biocides materials and other agricultural operations Figure 6.

Table 6: Four groups comprising the Contamination factors

Groups	**CF Value**	**contamination factor of Lagoon sediment samples**
A.	CF< 1	Low contamination.
B.	CF1 - 3	Moderate contamination.
C.	CF3- 6	Considerable contamination.
D.	CF>6	Very high contamination.

Table 7: Calculated contamination factor for total cadmium contentand cadmium fractionation in sediment samples of Lagoon.

Stations	**Total content**	**(EXC)**	**(CA)**	**(FM)**	**(OM)**	**(RES)**
		Exchangeable	**Carbonate bond**	**Fe-Mn hydroxide bond**	**Organic bond**	**Insoluble**
1	58	3.2	3.8	1.8	2.2	47
2	60	3.8	2.6	2.8	2.2	48.6
3	54	3.4	3.4	1.4	1.4	44.4
4	62	4	5	1.4	3.4	48.2
5	52	3.4	3	2.2	2.6	40.8
6	58	4.4	2.8	1.2	2	47.6
7	62	7	3.8	1.8	3	46.4
8	26	6.2	2	1.2	2.4	14.2
9	22	6	1.4	1.2	2	11.4
10	44	5.6	3.6	1	1.8	32
11	42	6	1.2	1.6	2	31.2
12	60	6.4	2.6	3.8	3.4	43.8
13	56	6	2.4	2.2	3.4	42
14	58	6.8	2.8	1.6	2.4	44.4
15	50	6.6	1.4	3.6	2.4	36
16	38	5.4	2.2	2.2	1.6	26.6
17	40	6.4	2.2	2.2	2.4	26.8
18	46	6	3.4	1.4	2.8	32.4
19	56	6.6	3.4	1.8	2.2	42
20	52	6.6	2.6	2	2	38.8
21	56	6	2.4	2.2	2.4	43
Average	49.7	5.63	2.71	1.94	2.39	37.03
Max	62	7	5	3.8	3.4	48.6
Min	22	3.2	1.2	1	1.4	11.4

The reducible fraction comprised organic fraction and Fe- Mn hydroxide fraction cadmium bound together. The sum of that two fractions bound ranges between 1.4 to 3.6 ppm with an average 2.2 ppm. It was noticed that reducible fraction content increases toward the south, the contractions and decreased contaminated spaces, confined to the south of the lagoon (Figure 7).

The residual fraction holds metals bound to mineral matrix. This fraction represents the metals that are slightest mobile and available to living organisms. This fraction ranges from 5.7 to 24.3 with an average 18.5ppm. The content increases due middle and eastern portion, generally, the residual fraction represent the dominant area of the lagoon (Figure 8).

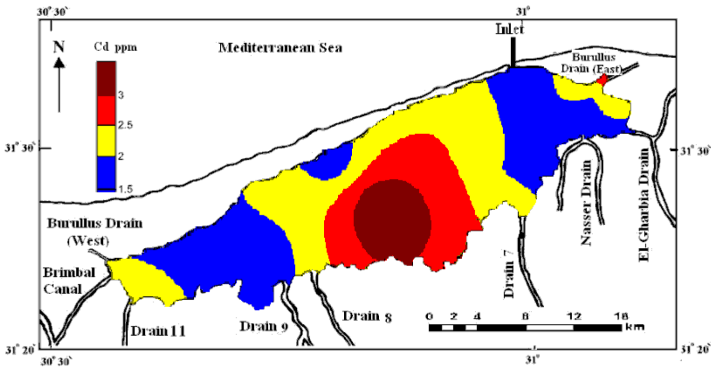

Figure (7): Geochemical map of reducible fraction content of cadmium (Sumorganic and Fe- Mn hydroxide fractions) in the studied lagoon sediment.

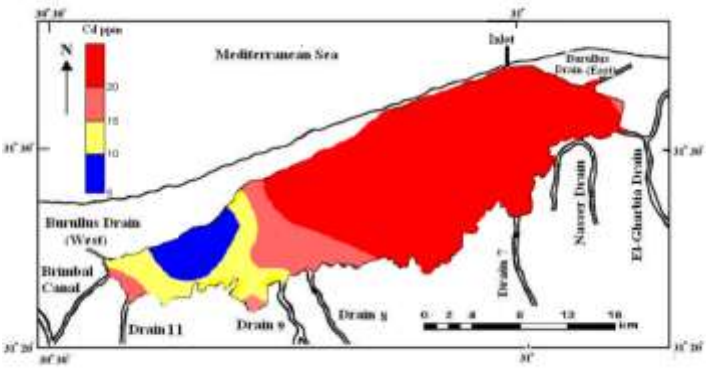

Figure (8): Geochemical map of residual content of cadmium in the studied lagoon sediment.

Table 8: seven classes comprising the geoaccumulation index

	Geoaccumulation index	
Classes	***Igeo* value**	**pollution type**
A.	< 0	Unpolluted
B.	0–1	Unpolluted to moderate
C.	1–2	Moderate
D.	2–3	Moderate to strong
E.	3–4	Strong
F.	4–5	Strong to extremely strong
G.	> 5	Extreme

Table 9: Calculatedgeoaccumulation index for total cadmium contentand cadmium fractionation in sediment samples of Lagoon.

Stations	Total content	(EXC) Exchangeable	(CA) Carbonate bond	(FM) Fe-Mn hydroxide bond	(OM) Organic bond	(RES) Insoluble
1	1.9	0.6	0.7	0.4	0.5	1.8
2	1.9	0.7	0.5	0.6	0.5	1.8
3	1.9	0.7	0.7	0.3	0.3	1.8
4	1.9	0.7	0.8	0.3	0.7	1.8
5	1.8	0.7	0.6	0.5	0.5	1.7
6	1.9	0.8	0.6	0.2	0.4	1.8
7	1.9	1	0.7	0.4	0.6	1.8

8	1.5	0.9	0.4	0.2	0.5	1.3
9	1.5	0.9	0.3	0.2	0.4	1.2
10	1.8	0.9	0.7	0.1	0.4	1.6
11	1.7	0.9	0.2	0.3	0.4	1.6
12	1.9	0.9	0.5	0.7	0.7	1.8
13	1.9	0.9	0.5	0.5	0.7	1.7
14	1.9	1	0.6	0.3	0.5	1.8
15	1.8	0.9	0.3	0.7	0.5	1.7
16	1.7	0.9	0.5	0.5	0.3	1.5
17	1.7	0.9	0.5	0.5	0.5	1.6
18	1.8	0.9	0.7	0.3	0.6	1.6
19	1.9	0.9	0.7	0.4	0.5	1.7
20	1.8	0.9	0.5	0.4	0.4	1.7
21	1.9	0.9	0.5	0.5	0.5	1.8
Average	1.8	0.9	0.6	0.4	0.5	1.7
Max	1.9	1	0.8	0.7	0.7	1.8
Min	1.5	0.6	0.2	0.1	0.3	1.2

Generally, on the average, Cd association with the various fractions followed the decreasing order: residual fraction> exchangeable fraction > carbonate fraction > organic fraction>Fe- Mn hydroxide fraction (Figure 9).

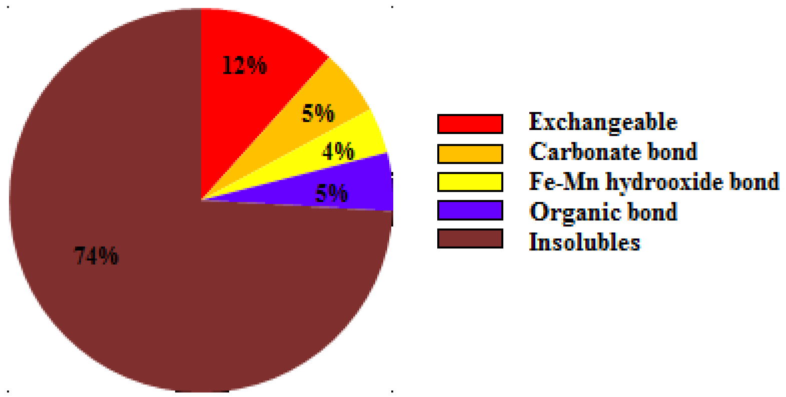

Figure 9: Distribution of Cd in various chemical fractions.

The pollution index:

The degree of pollution of the Brullus lake bottom sediments can be calculated by normalizing the metals concentration to their background value is that of average shale, obtained by McLennan and Taylor[22].

Risk assessment code (RAC)

The risk assessment code (RAC) fundamentally applies to the bioavailable speciation (sum of exchangeable and carbonate bound fractions) for assessing the availability of metals in sediments (Table 5). These code (RAC) suggested byPerin[36]etal.If a metal content in these fractions less than 1% of the total metal it will be considered safe for the environment. On the lagoon bottom sediment releasing in the same fractions more than 17% of the total metal content so, the metal species posed medium risk and can easily enter into the food chain.

Contamination Factor (CF)

The level of contamination can be expressed by the contamination factor (CF), Hökanson[37] classified the contamination into four groups Table 6.It was calculated as follows:

CF= (Metal content in the sediment) / (Metal content in natural reference sediment)

Studied heavy metals background values obtained by McLennan and Taylor [22]. The values of contamination factor (**CF**) are shown in Table 7. Very high contamination was showed for cadmium and residual fractionation step recorded at all stations of the lake, while bioavailable content comprise exchangeable and carbonate fractions tend to be moderate to considerable contamination Factor. Contamination factor for reducible fractions content of cadmium in the studied lagoon sediment showed moderate CF.

Geoaccumulation index (I_{geo})

The geoaccumulation index (I_{geo}) was originally defined by Müller[38]. It enables an assessment of the enrichment degree by considering the anthropogenic pollution, the geochemical background values, and the effect of natural digenesis. The I_{geo} was computed using the following equation:

$$I_{geo} = \log 2\ (C_n / 1.5 \times B_n)$$

Where Cn is the measured concentration of the element n in sediment sample, and Bn is the geochemical background value of the element n according to McLennan and Taylor[22]. A constant of 1.5 is used due to a given metal fluctuations in the soils as well as some very small anthropogenic influences Loska[39] et al. The geo-accumulation index consists of seven classes or grades (Table 8), and the highest class (six) reflects 100 fold enrichment above the background values Förstner[40] et al.

The geoaccumulation index values for the studied cadmium metal concentrations in bottom sediment lagoon are listed in Table 9. The calculated geoaccumulation index for steps 1, 2, 3 and 4 indicates that the lagoon unpolluted with cadmium fractionation, while geoaccumulation index values of cadmium showed moderate polluted categories for cadmium content and residual fractionation.

Conclusions:

Brullus lagoon is one of four shallow coastal water bodies in north of Egyptian Delta. It receives huge quantity of agricultural, industrial, municipal and domestic wastewater, in addition to navigation and fishing activities.

The concentration of cadmium in lagoon water have concentrations higher than those in Mediterranean Sea water, which indicate the influence of the evaporation on the lake water. The concentrations of cadmium metal increase toward the northeastern area of the lagoon.

The distribution of cadmium metal in surficial bottom sediments of Brullus lagoon revealed that the agricultural pollution represents by El-Maksaba and Mastarouh villages at northern area of the Lagoon, and the industrial district represented by El-Gharbia drain which throughout its waste water directly to the Lagoon containing the industrial wastes of textile factories located at El-Mahalla El-Kobra, also, industrial wastes as electroplating and synthetic fiber production, also the fishing boats seem to be main source of pollution. Among all the fractions for cadmium, the reducible fraction was the most abundant pool, followed by exchangeable, carbonate, organic and finally Fe- Mn hydroxide fraction .According to contamination factors and geoaccumulation factors relative to average earth's crust revealed that the lake sediments are markedly Very high cadmium contamination was recorded at all stations.

Following this study, we recommend following up on the variables related to the quality of water and heavy elements through systematic observation with perfect lagoon management to maintain this water system from deterioration and preserve fish wealth safe for human use.

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Reference

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