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ION ACTIVITIES OF SOME TRACE METALS IN AQUATIC ENVIRONMENTS

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ABSTRACT

In Egypt, Rosetta branch, one of the two main branches of the Nile River, is impacted by several agricultural drains and industrial companies at Kafr El-Zayat City which potentially affect and deteriorate its quality of water. In this study water and sediment samples were collected from twelve different locations in Kafr-El Zyat area. Analytical values including temperature, pH, total concentrations of trace metals, major anions and cations were inserted into the *Visual MINTEQ* geochemical speciation model to calculate various ionic activities of (Mn, Fe, Cu and Al). The results show that the studied metal in the collected samples are controlled by different solid phases. Mn activity is controlled by $MnCO_3(s)$, Cu is controlled by CuO(s) while Fe is controlled by $Fe(OH)_3$ and Al is controlled by $Al(OH)_3$ amorphous and gibssite. The study reveals that, identifying ionic activities is of particular importance as pollutants affect the water environment by the chemical behavior of the ionic species than by total concentrations. Finally the adverse effects of highly soluble metals are important in water chemistry because their inherent toxicities are related to the bioavailability.

Introduction:

The aquatic chemistry of trace metals in the natural environment depend upon the distribution dynamics of these metals and on the type of interaction between the metal and their aquatic habitat. The effect of pollutants on organisms in the aquatic environment is determined by the positional and bio availability of pollutants. The bio available fraction refers to that portion of the total amount of a pollutant present in a system which is potentially available for uptake by the organisms and positional availability **Grobler, D. C. and Davies, E.1979.**

Gun et al, 1988 stated that the trace metals are considered as contaminations of terrestrial and aquatic systems because of their persistence and toxicity at low concentrations. There is considerable evidence that the physicochemical form in

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which they are present in waters, sediments and in soils markedly influences the bioavailability and toxicity of such metals. El-Sanafawy (1997) stated that, Elements, both metallic and non metallic, may occur in one of several oxidation states and in soluble complexes with different organic and inorganic ligands.

Determination of collective parameters and total elemental composition is inadequate for identifying the mechanisms that control the composition of natural waters and indeed the soil solution. The chemical behavior of the elements in natural systems depends on their chemical speciation. The aim of this study is to estimate the total concentrations of anions and cations and the ion activities by using speciation model for both water and sediment samples in Kafr El-Zyat city.

Materials and Methods

Selected Sites

Water and sediment samples were collected in December 2005 from fifteen different sites at Kafr-El Zyate City.These selected sites are shown in the map (Fig1).

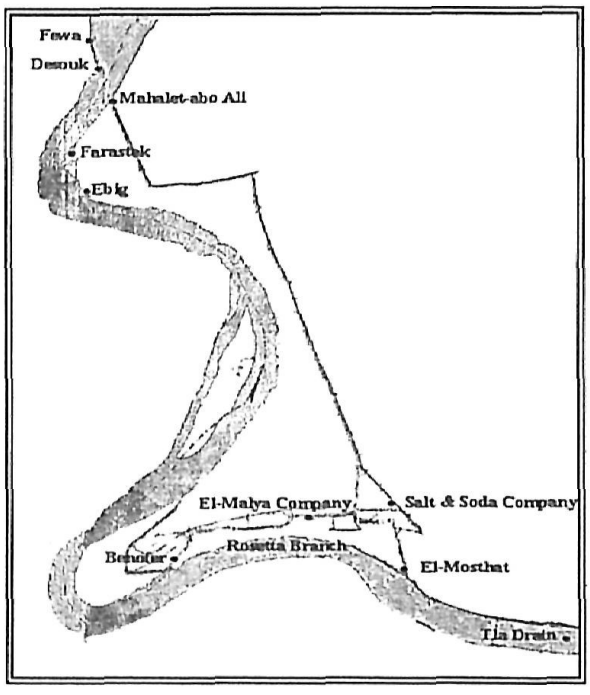

Figure (1): Map of the studied area

Sampling

Water samples were collected in one liter polyethylene container using auto samplers. Containers were rinsed using tap water, distilled water and finally by the site water before sample collection.

Samples were preserved following the procedures outlined by the **American Public Health Association "APHA" (1992)**, and transported to the laboratory in ice boxes. At each corresponding water sampling site, sediment samples were also collected using hand corer. Samples were stored in polyethylene bags *and* then transported to the laboratory in ice tanks within few hours from the collection.

Sample Analysis

Water Analysis

Field Analysis

Temperature, dissolved oxygen (DO), pH, and electrical conductivity (EC) were determined in-situ using a multi-probe system, model Hydro lab-Surveyor.

Laboratory Analysis

Total alkalinity values reported determined by electrometric titration of a sample aliquot using a standard solution of (0.02N H_2SO_4). The inflection points determined by pH-meter were in micro equivalents per liter. Trace metals and major cations were analyzed using Inductively Coupled Plasma-Optical Emission Spectroscopy (ICP-OES) with ultrasonic nebulizer (USN), Perkin Elmer Optima 3000, USA in clear filtrates obtained by 0.45 μm -pore-diameter filter paper. Major anions concentrations were determined using Ion Chromatography (IC), model DX-600 chromatography system.

Sediment Analysis

Collected sediment samples were air dried for 24 hours then crashed, extracted with water at a sediment : water ratio of 1: 2 and shaken for 1 day in a shaker model Rumo 3015. Suspensions were filtered using 0.45 μm-pore-diameter filter papers for analysis as indicated in water samples.

Visual MINTEQ Model

Analytical values including temperature, pH, and total concentration of trace metals, major anions and cations were inserted in the *Visual MINTEQ* geochemical speciation model, US EPA to calculate various ionic activities. The ionic strength (I) was not fixed and left to be calculated by the iterative program. Temperature was set at 25^0C for sediment samples. Ionic Activities of Al^{+3}, Cu^{+2}, Mn^{2+} and Fe^{3+} as well as their important ionic complexes were obtained from the *Visual MINTEQ* output files. Total concentrations and ion activities of these metals were plotted on the solubility diagrams similar to those developed by **Lindsay 1979, Tawfic 1990, Tawfic &Lindsay 1995.**

Results and Discussion

1. Physicochemical parameters

1.1. pH

The results of the pH value of surface water samples ranged (7.5-7.84) which means that all water samples are alkaline and the values of sediment extracts for the same samples are ranged from (7.1-7.9). Sediment pH is affected by the changes in redox potential that occur in sediments that become waterlogged periodically. Reducing conditions generally cause a pH increase and oxidation brings about a decrease **(Alloway, 1990).**

1.2. Electrical Conductivity

The results show that, the electrical conductivity (EC) for water and sediment extracts ranged (503 - 1520 µS/cm) and (625- 5050 µS/cm) respectively. EC values for sediments are higher than that for water samples for all locations .Which due to higher concentrations of cations and anions in sediment extracts than in water samples. Results are shown in appendix 1.

2. Chemical parameters

2.1. Major anions

In all the studied sampling locations, the fluoride concentration is ranged between (0.203 - 0.575 mg/l) in water samples, and between (0.001 - 0.594 mg/l) in sediment extracts. Chloride concentrations in water samples ranged between (35.8 -

178.7 mg/l), and between (13.2-1005.4 mg/l) for sediment extracts. Nitrate concentrations ranged from (1.19 - 31.8 mg/l) in water samples, and between (0 - 332.4 mg/l) in sediment extracts. Concentration of sulfate in water samples is ranged between (41.7 - 252.3 mg/l), and between (146.4 - 8204.2 mg/l) in sediment extracts. Complete results are shown in appendix 1.

2.2. Major cations

Sodium concentrations ranged between (40 - 190 mg/l) in water samples, and between (65 - 640 mg/l) in sediment extracts. Calcium concentrations ranged between (36.3-132 mg/l) in water samples, and (75.9 - 1280 mg/l) in sediment extracts. Magnesium concentrations ranged between (15 - 45.5 mg/l) in water samples and between (29 - 472mg/l) in sediment extracts. Potassium concentrations ranged between (9.1- 13.1mg/l), in water samples and between (15 - 52.2 mg/l) in sediment extracts. The results of major cations revealed that the water samples were lower in concentration than sediment extracts for all locations.

3.3. Trace metals ion activities

3.3.1. Iron

As it is shown from the appendix 2 the concentration of iron range between (0.017-0.414 mg/l) in water samples and between (0.012 - 0.618 mg/l) in sediment extract samples.

Total concentrations and calculated activities of iron were plotted on Figure (2), for iron The line drawn is $Fe(OH)_3$ (*amorphous*) using the following equations

$$Fe(OH)_3 \text{ (amorphous)} + 3H^+ \rightleftharpoons Fe^{+3} + 3H_2O \qquad \log K^0 = 3.54$$

$$\text{Log } (Fe)^{+3} = 3.54 - 3pH$$

By plotting these data, it was shown that iron ion activities estimated by the model and the logarithmic values of iron total concentrations all fall above $Fe(OH)_3$(amorphous) line which suggests that all samples are supersaturated with respect to all iron species lines including this line. **Lindsay 1979**

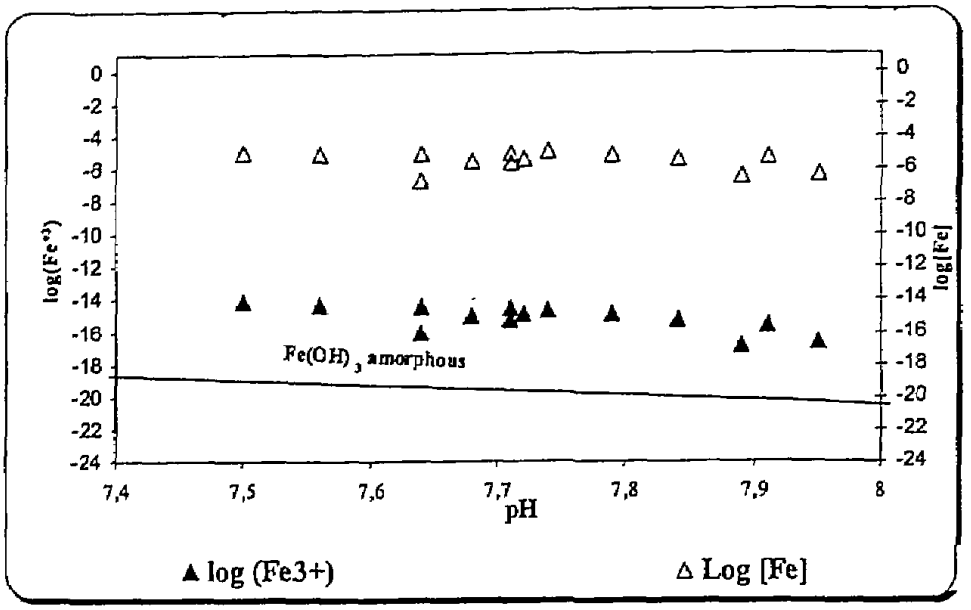

Figure (2): Total conc. and activity of Fe^{3+} calculated from Visual MINTEQ showing data points of Fe^{3+} are supersaturated with respect to Fe $(OH)_3$ (amorphous) lines

3.3.2. Manganese

From data in appendix 2 it shows that the total concentration of manganese in water range between (0.01 - 0.309 mg/l), and for sediment samples the total concentration of manganese range between (0.148 - 18.50 mg/l), this means that the total concentration of manganese in sediment samples is much greater than in water sample.

Total concentration of manganese were plotted on Figure (3) drawn by **Lindsay 1979**, using equations

$$MnCO_3 + 2H^+ \rightleftharpoons Mn^{+2} + CO_2 + H_2O \qquad \log K^0 = 8.08$$

At $\log CO_2 = -2.5$ $\log (Mn^{+2}) = 10.58 - 2pH$

At $\log CO_2 = -1.5$ $\log (Mn^{+2}) = 9.58 - 2pH$

At $\log CO_2 = -0.57$ $\log (Mn^{+2}) = 8.08 - 2pH$

By plotting these data, it is shown that manganese ion activities estimated by the model are all around $MnCO_3$ line which suggests that $MnCO_3$ is most probably the

solid phase that control manganese activities. The data also indicates that (Mn) decreases with the increase in CO_2 (g) partial pressure.

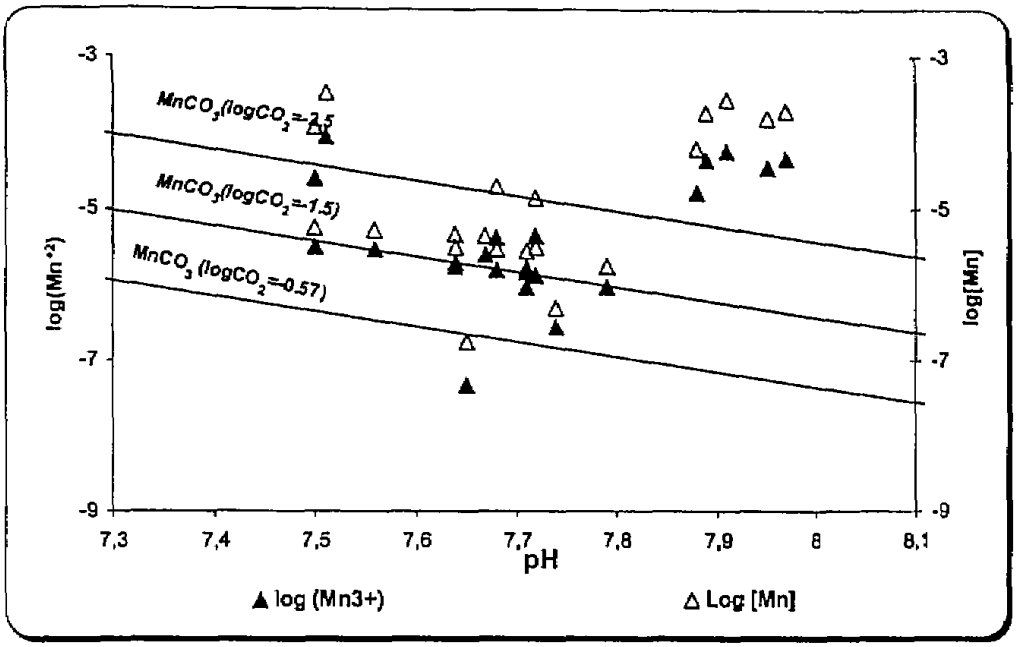

Figure (3): Total conc. and activity of Mn^{2+} calculated from Visual MINTEQ showing data points of Mn^{2+} fall between $MnCO_3$ lines

3.3.3 Copper:

From data in appendix 2 it shows that the total copper concentrations in water samples ranged between (0.002 -0.011 mg/l), and in sediment between (0.006 - 0.087 mg/l)

The total concentration and activity of Cu^{+2} which calculated from Visual MINTEQ were plotted on Figure (4) drawn by **Lindsay 1979** according to the following equilibrium reaction:-

$$CuO(\text{ tenorite}) + 2H^+ \rightleftharpoons Cu^{+2} + H_2O \qquad \log K^0 = 7.66$$

$$\text{Log } (Cu^{+2}) = 7.66 - 2pH$$

By plotting these data, it is shown that copper ion activities estimated by the model are all around CuO (tenorite) line which suggests that tenorite is the solid phase controls copper activities.

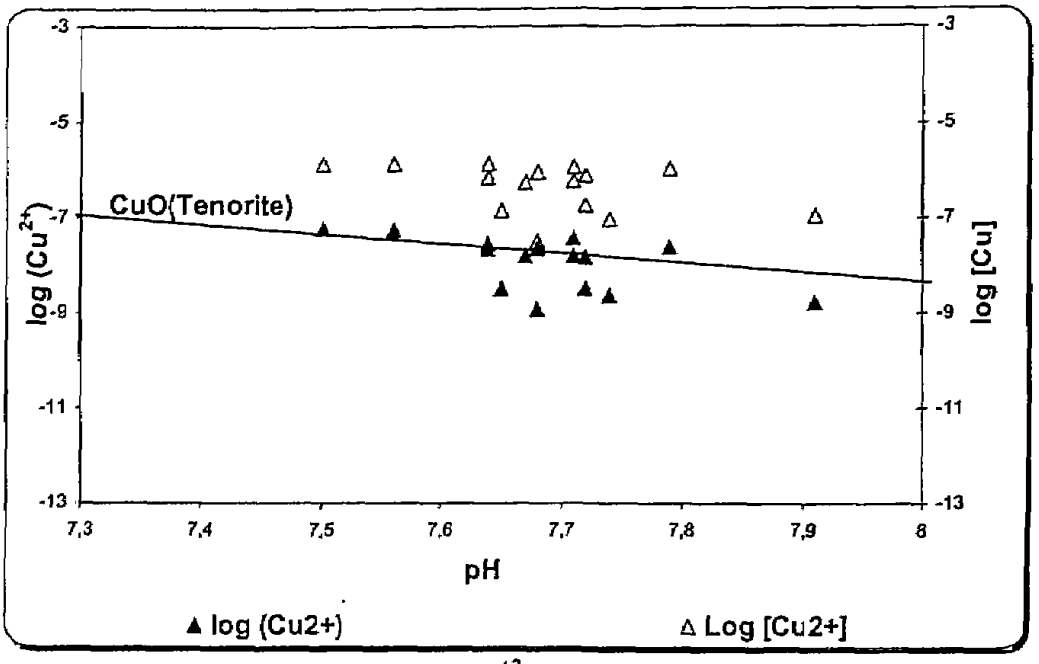

Figure (4): Total conc. and activity of Cu^{+2} calculated from Visual MINTEQ showing data points of Cu^{+2} fall around tenorite line.

3.3.4 Aluminum

From the results in appendix 2 it shows that the total concentrations of aluminum in water samples ranged between, (0.011 - 0.54 mg/l) and between (0.042 - 0.959 mg/l) for sediment extract.

The total concentration of aluminum are plotted on Figure (5) which was drawn by **Lindsay, 1979**, using equations

$$Al(OH)_3 \text{ (amorphous)} + 3H \rightleftharpoons Al^{+3} + 3H_2O \quad \log K^0 = 7.66$$

$$\text{Log } (Al^{+3}) = 9.66 - 3pH$$

$$\gamma\text{-}Al(OH)_3 \text{(Gibbsite)} + 3H^+ \rightleftharpoons Al^{+3} + 3H_2O \quad \log K^0 = 8.04$$

$$\text{Log } (Al^{+3}) = 8.04 - 3pH$$

By plotting these data in Figure (5), it is shown that aluminum ion activities are all between γ-Al(OH)$_3$(Gibbsite) line and Al(OH)$_3$ (amorphous) line which show that gibbsite may be the solid phase that control aluminum activities.

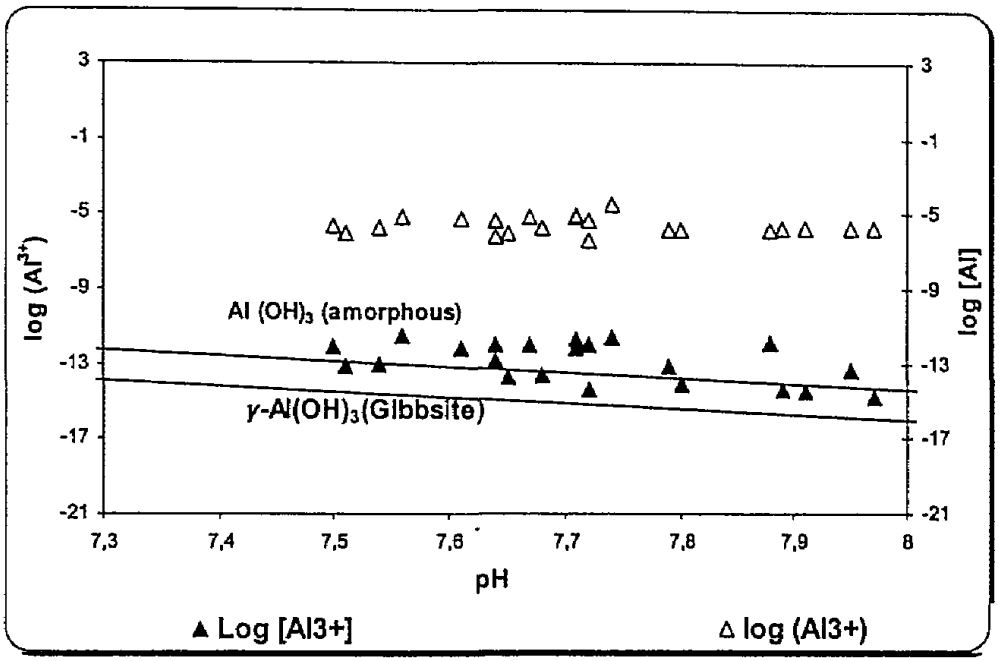

Figure (5): Total conc. and ion activity of Al^{+3} calculated from Visual MINTEQ Showing that all data points lay between Al (OH) $_3$ (amorphous) and (Gibbsite) line

Conclusion:

It is now becoming evidence that metal speciation into different fractions is the most reliable criterion for quantifying the potential effect of contamination of sediments by trace metals. The data indicated that the bioavailability of trace metals in aquatic system is best explained by activities rather than concentrations. The results also show that the metal speciation give an insight to the behavior of the chemical forms that is present in solution.

Visual MINTEQ model suggest that iron activity is supersaturated with respect to Fe(OH)$_3$ (amorphous), $MnCO_3$ may be the solid phase that control manganese activity, Copper activity is controlled by Tenorite CuO(s), while Al is controlled by Gibbsite γ-Al(OH)$_3$ and Al(OH)$_3$ (amorphous).

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Appendix 1, Results of physical and chemical analysis of water and sediment extract samples

Sample Code	Sample Description	pH	EC (µS/cm)	K^+	Mg^{2+}	Ca^{+2}	Na^+	SO_4^{2-}	PO_4^{3-}	NO_3^-	Cl^-	F^-
R1 (Rosetta beforeTala Drain)	Water Sample	7.71	504	10.9	16.7	39.7	40.8	41.721	<0.2	2.078	37.26	0.273
	Sediment extract	7.65	3610	41.3	195	691	471	2157.88	<0.2	1.428	60.2	0.148
R2 (Tala mix Rosetta)	Water Sample	7.72	1510	12	44.5	82.8	189	250.085	<0.2	31.34	177.397	0.291
	Sediment extract	7.54	4850	38.7	408	1110	378	8204.262	<0.2	8.78	394.77	0.56
R3 (Rosetta after Tala Drain)	Water Sample	7.64	1520	13.1	45.5	85.4	190	252.346	<0.2	31.864	178.686	0.381
	Sediment extract	7.8	4540	30	310	520	210	3320	<0.2	4.2	210	0.59
R4 (El-mosatahat)	Water Sample	7.79	522	10.4	15.5	37.2	50.5	45.301	<0.2	1.577	39.014	0.295
	Sediment extract	7.72	2100	27.1	92.2	476	65	1115.88	<0.2	2.75	17.49	0.176
R5 (Salt & Soda Co.effluents)	Water Sample	7.5	655	10.1	16.8	42.8	69	44.462	<0.2	2.833	95.714	0.279
	Sediment extract	7.91	4370	40.5	283	1110	192	30609.7	<0.2	<0.2	62.95	0.5
R6 (El-Malia Co.)	Water Sample	7.56	651	10.6	17	42.9	67.8	44.379	<0.2	2.918	97.153	0.355
	Sediment extract	7.95	3400	40.5	175	884	128	2683.23	<0.2	5.212	38.7	0.583
R7(Rosetta at Benofer)	Water Sample	7.84	525	10	16.1	38	50	44.283	<0.2	1.712	42.132	0.203
	Sediment extract	7.89	3980	44.3	240	1070	163	2839.85	<0.2	0.504	37.45	0.005
R8(Rosetta at Ebig)	Water Sample	7.74	517	10.8	16.2	37.7	49	43.854	<0.2	1.285	37.667	0.305
	Sediment extract	7.97	3970	47.2	262	1070	145	3321.38	<0.2	<0.2	29.15	0.132
R9 (Rosetta at Farastek)	Water Sample	7.71	516	10.6	15.8	36.3	49	43.751	<0.2	1.199	41.202	0.372
	Sediment extract	7.68	4970	25.5	288	860	507	4120	<0.2	1.4	40.2	0.11
R10 (Rosetta at Mahalet abo ali)	Water Sample	7.68	533	9.93	15.8	38.7	51.6	43.428	<0.2	1.602	38.096	0.575
	Sediment extract	7.88	2670	29.2	165	792	88.2	2380.767	<0.2	6.094	16.349	0.224
R11(Rosetta at Desouk)	Water Sample	7.64	522	9.1	15.1	37.7	50	44.263	<0.2	1.477	37.453	0.396
	Sediment extract	7.51	2910	34.3	164	638	137	2225.59	<0.2	6.503	28.681	0.29
R12 (Rosetta at Fewa)	Water Sample	7.67	503	9.83	15	36.5	49.2	42.91	<0.2	1.393	35.823	0.376
	Sediment extract	7.92	625	15	29	75.9	93.5	192.653	<0.2	332.47	31.156	0.54

Appendix 2, Results of total and ion activities of trace metals in water and sediment extract samples

Sample Code	Sample Description	[Fe] mg/l	Log[Fe] mol/l	Log(Fe^{+3}) mol/l	[Mn] mg/l	Log[Mn] mol/l	Log(Mn^{+2}) mol/l
R1 (Rosetta beforeTala Drain)	Water Sample	<0.01	–	–	0.01	-6,739	-7,323
	Sediment extract	0.421	-5,122	-14,579	0.148	-5.56	-5,816
R2 (Tala mix Rosetta)	Water Sample	<0.01	–	–	—	–	–
	Sediment extract	0.206	-54,331	-14,923	0.173	-5,502	-5,882
R3(Rosetta after Tala Drain)	Water Sample	<0.01	–	–	—	–	–
	Sediment extract	0.452	-5,091	-14,465	0.26	-5,342	-5,701
R4 (El-mosatahat)	Water Sample	<0.01	–	–	0.1	-5,739	-6,013
	Sediment extract	0.4	-5,144	-14,853	0.771	-4,852	-5,362
R5 (Salt & Soda Co.)	Water Sample	0.272	-5,312	-15,533	0.309	-5,249	-5,491
	Sediment extract	0.618	-4,955	-14,124	15.1	-3,561	-4,228
R6 (El-Malla Co.)	Water Sample	0.024	-6,366	-16,667	0.284	-5,286	-5,529
	Sediment extract	0.498	-5,049	-14,286	8.53	-3,808	-4,438
R7 (Rosetta at Benofer)	Water Sample	0.017	-6,516	-16,866	—	–	–
	Sediment extract	0.191	-5,465	-15,261	10.4	-3,723	-4,343
R8 (Rosetta at Ebig)	Water Sample	0.017	-6,516	-16,866	0.028	-6,293	-6,558
	Sediment extract	0.543	-5,011	-14,651	11	-3,698	-4,337
R9 (Rosetta at Farastek)	Water Sample	<0.01	–	–	0.092	-5,776	-6,015
	Sediment extract	0.113	-5,693	-15,238	1.09	-4,702	-5,372
R10 (Rosetta at Mahalet abo ali)	Water Sample	<0.01	–	–	0.165	-5,522	-5.79
	Sediment extract	0.179	-5,493	-14,959	3.42	-4,206	-4,777
R11 (Rosetta at Desouk)	Water Sample	<0.01	–	–	0.17	-5,509	-5,754
	Sediment extract	0.012	-6.66	-16,036	18.5	-3,472	-4,055
R12 (Rosetta at Fewa)	Water Sample	<0.01	–	–	–	–	–
	Sediment extract	<0.01	–	–	0.264	-5,348	-5,602

Sample Code	Sample Description	[Cu] mg/l	Log[Cu] mol/l	Log(Cu^{+2}) mol/l	[Al] mg/l	Log [Al] mol/l	Log(Al^{+3}) mol/l
R1 (Rosetta beforeTala Drain)	Water Sample	0.009	-6,849	-8.48	0.027	-5,599	-13,712
	Sediment extract	0.074	-5..934	-7,448	0.244	-50,436	-11,678
R2 (Tala mix Rosetta)	Water Sample	–	–	–	0.051	-5,723	-13,023
	Sediment extract	0.047	-6,131	-7,834	0.123	-5,341	-12,001
R3(Rosetta after Tala Drain)	Water Sample	–	–	–	0.042	-5,808	-14.12
	Sediment extract	0.085	-5,874	-7,528	0.109	-5,393	-11,945
R4(El-mosatahat)	Water Sample	0.011	-6,762	-8.49	0.011	-6,389	-14,365
	Sediment extract	0.066	-5,984	-7,634	0.042	-5,808	-13,092
R5 (Salt & Soda Co.effluents)	Water Sample	0.007	-6,958	-8.78	0.054	-5,699	-14,452
	Sediment extract	0.087	-5,864	-7,221	0.068	-5,599	-12,034
R6 (El-Malla Co.)	Water Sample	–	–	–	0.045	-5,778	-13,279
	Sediment extract	0.082	-5,889	-7,261	0.198	-5,134	-11,468
R7(Rosetta at Benofer)	Water Sample	–	–	–	–	–	–
	Sediment extract	–	–	–	0.049	-5,741	-14,411
R8(Rosetta at Ebig)	Water Sample	–	–	–	0.05	-5,732	-14,722
	Sediment extract	0.006	-7,024	-8.63	0.959	-4,449	-11,603
R9 (Rosetta at Farastek)	Water Sample	0.002	-7,502	-8,916	0.046	-5,768	-13,609
	Sediment extract	0.037	-6,235	-7,802	0.202	-5,126	-12,172
R10 (Rosetta at Mahalet abo ali)	Water Sample	–	–	–	0.042	-5,808	-11,886
	Sediment extract	0.056	-6,055	-7,651	0.053	-5,707	-12,545
R11(Rosetta at Desouk)	Water Sample	–	–	–	0.015	-6,255	-12,866
	Sediment extract	0.042	-6,179	-7,666	0.027	-5,999	-13,155
R12 (Rosetta at Fewa)	Water Sample	–	–	–	–	–	–
	Sediment extract	0.034	-6,272	-7.81	0.188	-5,157	-12,022

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   g) Conclusions h) Acknowledgements i) References
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4. The meaning of all symbols should be given immediately after the equation in which they are first used.
5. Equations should be numbered serially on the right-hand side and in parantheses.
6. In chemical formulas the valence of ions should be given as e.g. :
   Ca^{2+} and CO_3^{2-} rather than Ca^{++} and CO_3^{--}
7. Isotope number should precede the symbol e.g. : ^{18}O.

REFERENCES

1. All references to publications made in the text should be presented in a list of references following after the text. The manuscript should be carefully checked to ensure that the spelling of author's names and dates are exactly the same in the text as in the reference list.
2. References in the text should be arranged chronologically. The list of references should be arranged alphabetically by authors' names, and chronologically per author.
3. The following system should be used for arranging references:
   a) For Periodicals: Lamb,H.H., Climate Engineering, 7,87-95 (1971).
   b) For Books : Vanmeurs,A.P., Petroleum Economics, Elsevier (1972).
4. Periodical names can be given in full or abbreviated.

FOOTNOTES

1. Footnotes should only be used if absolutely essential.
2. If used, footnotes should be indicated by astrisks and kept as short as possible.

ملحوظة : يرفق مع البحث البيانات الاتيه فى ورقه منفصلـــــــــه :-

١) اسم الباحث الرئيسى .

٢) اسم وعنوان وتليفون جهه البحث

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