JOINT INSTITUTE FOR NUCLEAR RESEARCH

Frank Laboratory of Neutron Physics

FINAL REPORT ON THE

SUMMER STUDENT PROGRAM

EPITHERMAL NEUTRON ACTIVATION ANALYSIS OF SOME SCLERACTINIAN RED SEA CORALS

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Abstract. Five corals belonging to common scleractinian reef-building species collected from Suez Gulf, Egypt were analyzed using neutron activation analysis (NAA). Heavy metals and trace elements are measured for samples in three phases raw, clean and precipitate in order to assess the pollution level in the studied site. The analysis state that corals are good indicators for historical pollution of human activities during their lifetime. Iron has the highest value of metal concentration among twenty nine analyzed elements in all phases and higher than the concentration of the same elements in other sites along Red sea. Mn concentration has higher value in comparison with other sites in the Red sea of Eygpt. Distinct observation related of availability of uranium in the studied area.

Introduction. Corals display important components of the sea ecosystems. Due to the biological mineralization corals consider as chronometer (Al-Horani et al., 2003; Knutson et al., 1972). The most important advantage of using corals for pollution studies is that they settled in the same place through their life, which is relatively long. Also scleractinian corals have natural properties like, the accumulation the pollutant without being killed by the relatively high levels encountered in the marine environment, and therefore representative of the study sites,(Abdelbaset S. El-Sorogy • Mohamed A. Mohamed • Hamdy E. Nou). The main types of pollutants under consideration heavy metals, spilled petroleum from marine traffic and anthropogenic organics such as herbicides and pesticides. Suez Gulf has very large scale of maritime transport during the year, so it expected that it suffers from extensive pollution due to last causes and we aimed from this study to assess the pollution level of this site and introduce pollution scheme for a specific time period.

Methods.

Study area

The study site Suez Gulf (29°52'N and 32°28'E) is located in the northern end of the Red Sea, which bifurcates into the Sinai Peninsula, creating the Gulf of Suez in the west and the Gulf of Aqaba to the east. (See Fig.1). The length of the gulf, from its mouth at the Strait of Jubal to its head at the city of Suez, is 195 miles (314 km), and it varies in width from 12 to 20 miles (19 to 32 km). The gulf is connected to the Mediterranean Sea by the Suez Canal (north) and is an essential transportation route. Settlements along the gulf are confined to a few fishing and mining villages.

Figure 1, SuezGulf

Sample collection.

Five samples of Scleractinian corals of different common species Fig. (2) Were collected at a depth of around 2 m from the convergent areas that include modern constructions and human activities. The samples were washed by sea water and put into plastic bags until they were transferred to laboratory for measurements.

Figure 2. Scleractinian corals

Samples preparation for epithermal neutron activation analysis (ENAA).

Neutron Activation Analysis (NAA) is a sensitive analytical technique useful for performing both qualitative and quantitative multi-element analysis of major, minor, and trace elements in samples from almost every conceivable field of scientific or technical interest. For many elements and applications, NAA offers sensitivities that are superior to those attainable by other methods, on the order of parts per billion or better. In addition, because of its accuracy and reliability, NAA is generally recognized as the "referee method" of choice when new procedures are being developed or when other methods yield results that do not agree. Worldwide application of NAA is so widespread it is estimated that approximately 100,000 samples undergo analysis each year.

A sufficient portion of each sample was cut from the center of the coral using the diamond saw for NAA, The preparation of samples was conducted in three stages. Each portion was divided into two parts. The first part was taken as the raw phase; crushed by mortar to small pieces, then powdered by center fusion homogenizer. The second part was immersed in acetic acid (1:1) for 24 h, and then rinsed by distilled water, and subsequently the cleaned samples and their residues were dried at 40 C^0 for 3-4 days. After drying, centrifuging, and weighing the samples of 0.3 mg from each part were wrapped in high purity aluminum foil cups for determination of long-lived radionuclide and another 0.3 mg were wrapped in heat-sealed in polyethylene bags for short-lived radionuclides determination. NAA was carried out for the raw (unclean), cleaned and precipitate samples Fig. (3).

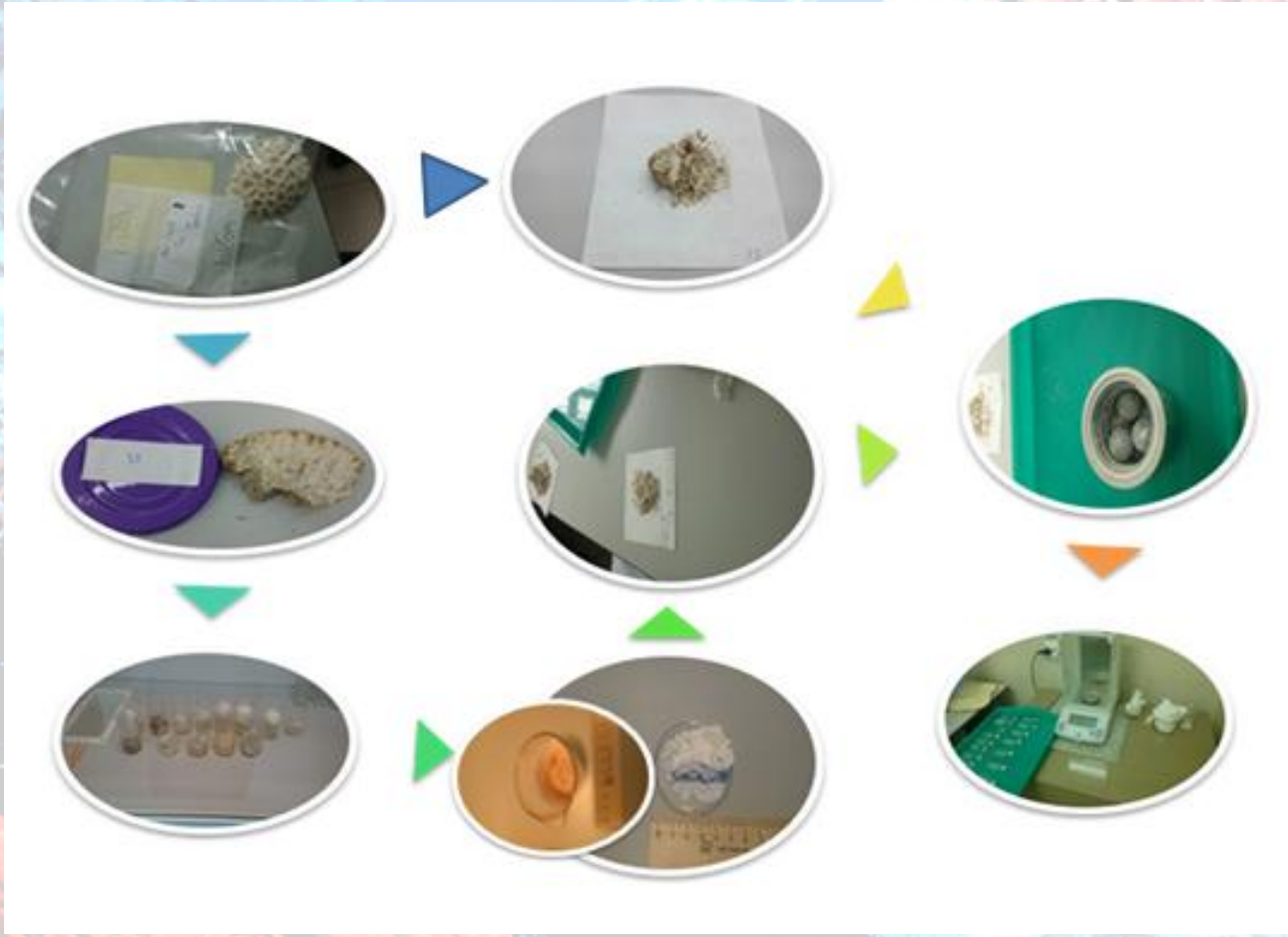


Figure 3. Cycle of sample preparation

Neutron activation analysis

For short irradiation coral samples of about 0.3 g were heat-sealed in polyethylene bags. For long irradiation samples of the same weight (about 0.3 g) were packed in aluminium cups.The NAA was performed at the pulsed fast reactor IBR-2 at the Frank Laboratory of Neutron Physics, Dubna, Russia. Two different irradiation times were used: 60 seconds (Ch 2 – for conventional NAA) and 100 hours (Ch1 – for epithermal NAA).Characteristics of neutron flux density in the channels equipped with the pneumatic system are given in Table 1.

Table 1.Flux parameters of irradiation positions

Irradiation position	$\Phi_{th} \cdot 10^{12}$, n cm^{-2} s^{-1} E=0 ÷ 0.55 eV	$\Phi_{th} \cdot 10^{12}$, n cm^{-2} s^{-1} E=0.55 ÷ 10^5 eV	$\Phi_{th} \cdot 10^{12}$, n cm^{-2} s^{-1} E=10^5 ÷ 25 10^6eV
Ch1 (Cd-screened)	0.023	3.3	4.2
Ch2	1.23	2.9	4.1

To determine short-lived isotopes samples were irradiated for 3 min. After irradiation two gamma-spectrometric measurements were performed; the first one for 5 minutes after 2–3 minutes of decay, and the second for 20 minutes after 9–10 minutes decay. Long-lived isotopes were determined after irradiation for 100 hours in the cadmium-screened Channel 1. After irradiation samples were re-packed into clean containers and measured after 4–5 and 20–23 days for 45 minutes and for 3 hours, respectively.

A total of 29 elements (Na, Mg, Al, Cl, Ca, Sc, Ti, V, Cr, Mn, Fe, Co, Zn, As, Br, Rb, Sr, Sb, I, Cs, Ba, La, Ce, Gd, Tb, Hf, Ta, Th, and U) were determined.

Table 2 lists selected peak energies for NAA and method of analysis. The gamma-spectra of the induced activity were analysed using software developed in Frank Laboratory of Neutron Physics (Pavlov et al., 2016).

Table 2. List of selected peak energies for NAA and method of analysis

Element	Isotope	Half life	Gamma peak (keV)	Method of analysis*
Na	^{24}Na	14.7 h	2753.6	3
Mg	^{27}Mg	9.5 m	1014.1	2
Al	^{28}Al	2.2 m	1778.9	1
Cl	^{38}Cl	37.2 m	2168.8	2
Ca	^{49}Ca	8.7 m	3084.4	2
Sc	^{46}Sc	83.8 d	889.2	4
Ti	^{51}Ti	5.8 m	320.1	1
V	^{52}V	3.8 m	1434.1	1
Cr	^{51}Cr	27.7 d	320.1	4
Mn	^{56}Mn	2.6 h	1810.7	2
Fe	^{59}Fe	44.5 d	1099.2	4
Co	^{60}Co	5.3 y	1173.1	4
Zn	^{65}Zn	244.0 d	1116.0	4

As	^{76}As	26.3 h	559.1	3
Br	^{82}Br	35.3 h	776.5	3
Rb	^{86}Rb	18.7 d	1076.6	4
Sr	^{85}Sr	64.8 d	514.0	4
Sb	^{124}Sb	60.2 d	1691.0	4
I	^{128}I	25.0 m	442.9	2
Cs	^{134}Cs	2.1 y	795.8	4
Ba	^{131}Ba	11.8 d	496.8	4
La	^{140}La	40.2 h	1596.5	3
Ce	^{141}Ce	32.5 d	145.4	4
Gd	^{153}Gd	241.6 d	103	4
Tb	^{160}Tb	72.3 d	879.4	4
Hf	^{181}Hf	42.4 d	482.0	4
Ta	^{182}Ta	114.4 d	1221.4	4
Th	^{233}Pa	27.0 d	312.0	4
U	^{239}Np	2.4 d	228.2	3

*Method 1: conventional NAA, measured after 2–3 min. of decay;
Method 2: conventional NAA, measured after 9–10 min. of decay;
Method 3: epithermal NAA, measured after 4–5 days of decay;
Method 4: epithermal NAA, measured after 20–23 days of decay.

Quality control

The QC of NAA results was ensured by simultaneous analysis of the examined samples and reference materials (2711, 1633, 667, 1633b, 1572 and 1547) ,(International Atomic Energy Agency), NIST standards.

Table 3. Comparison of measured and certified concentration values of reference material: 2711 (Montana Soil), 1633c (Coal fly ash), 667 (Estuarine sediment), 1633b (Coal fly ash), 1572 (Citrus Leaves), and 1547 (Peach leaves) for the neutron activation analysis

Elements	Certified value, mg/kg		Present work, mg/kg	
	Concentration, mg/kg	Uncertainty, %	Concentration, mg/kg	Uncertainty, %
Na	11400	2.6	11390.6	2.8
Mg	4320	1.9	4317.2	2.8
Al	150500	1.8	151172	3.1
Cl	360	5.3	360.7	8.4
Ca	15600	1.3	15661.4	5.4
Sc	37.6	1.6	37.6	2

Ti	7910	1.8	7908.9	5.3
V	295.7	1.2	296.1	3.6
Cr	258	2.3	258.2	5.7
Mn	131.8	1.3	132.1	5.3
Fe	44800	2.2	44818	4.6
Co	23	5.6	23	5.7
Zn	350	1.4	350	2.7
As	186.2	1.6	185.7	2.1
Br	99.7	2.5	99.7	2.7
Rb	117	0.5	117	5.4
Sr	245.3	0.3	244.9	6.5
Sb	19.4	9.3	19.4	9.6
I	1.84	1.6	1.83	10
Ba	1126	2.9	1130.5	4.3
Cs	9.39	2.3	9.39	2.8
La	87	3	87.3	3.2
Ce	56.7	4.4	56.7	5.3

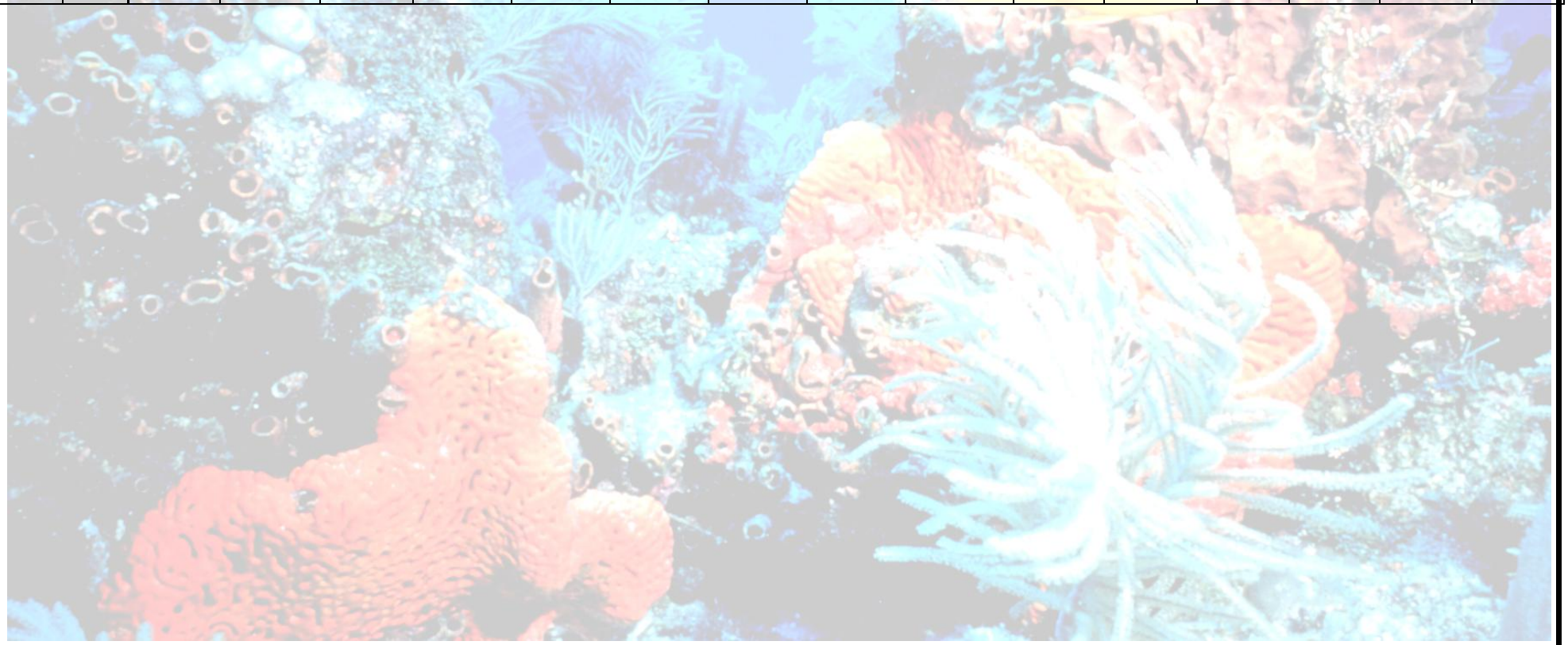
Gd	4.41	2.7	4.41	6.4
Tb	3.1	1.9	3.1	2.3
Hf	7.3	30	7.3	30
Ta	1.58	1.9	1.58	2.5
Th	10	5.0	9.9	5.2
U	9.25	4.8	9.23	4.9

Results of Neutron Activation Analysis

Table 4.Concentrations of elements (mg/kg) in cleaned corals collected in the Suez Gulf of Egypt

Element	sample	z-01	z-06	z-11	z-02	z-07	z-12	z-03	z-08	z-13	z-04	z-09	z-14	z-05	z-10	z-15
Na	mg/kg	3740 (3%)	3390(3%)	717(3%)	4780(3%)	1840(3%)	773(3%)	8490(3%)	4090(3%)	63400(3%)	1720(3%)	1190(3%)	1960(3%)	1230(3%)	864(3%)	8230(3%)
Mg	mg/kg	2230 (6%)	3500 (6%)	1230(6%)	1300 (6%)	5640 (6%)	4480 (6%)	5890 (6%)	2300(6%)	12200 (6%)	2350(6%)	1300(6%)	8060 (6%)	1320(6%)	1340 (6%)	5340 (6%)
Al	mg/kg	2350 (3%)	5100 (3%)	1050(3%)	1280 (3%)	5980 (3%)	3990 (3%)	1320 (3%)	608 (3%)	315 (19%)	487 (3%)	141 (3%)	10400 (4%)	503(3%)	561 (3%)	175 (9%)
Cl	mg/kg	1060 (9%)	196 (9%)	912 (9%)	2300 (9%)	291 (9%)	271 (9%)	11100 (9%)	998 (9%)	114000 (9%)	4030(9%)	976 (9%)	19100(9%)	1380(9%)	860 (9%)	35800 9%)
Ca	mg/kg	36% (6%)	28%(6%)	18 (6%)	32%(6%)	28(6%)	22%(6%)	32%(6%)	34%(6%)	13%(6%)	34%(6%)	42%(6%)	13%(6%)	33%(6%)	40%(6%)	14%(6%)
Sc	mg/kg	1.12 (3%)	4.51(3%)	0.994(3%)	0.956(3%)	4.71 (3%)	2.78 (3%)	0.453 (3%)	0.17 (3%)	0.153 (23%)	0.126 (3%)	0.0687 (3%)	0.119 (8%)	0.273 (3%)	0.269 (3%)	0.0708 (8%)
Ti	mg/kg	500 (11%)	850 (11%)	216 (11%)	212 (11%)	1140 (11%)	734 (11%)	673 (11%)	123(11%)		<170	79.6(11%)		<170	95.7 (11%)	
V	mg/kg	5.19 (5%)	9.63 (5%)	2.33 (5%)	3.04 (5%)	13.4 (5%)	9.48 (5%)	5.97 (5%)	2.13 (5%)	<9.32	1.69 (5%)	0.564 (5%)	5.07 (30%)	1.02 (5%)	1.08 (5%)	1.12(30%)
Cr	mg/kg		26.2 (8%)			37.6 (8%)	26.9 (5%)		12.9(8%)	<6.6	<7	18.6 (8%)		13 (15%)	14.3 (8%)	
Mn	mg/kg	90.9 (5%)	170 (5%)	53.9 (5%)	45.8 (5%)	173 (5%)	92.3 (5%)	41.2 (5%)	7.98 (5%)	35.8 (5%)	9.74 (5%)	4 (5%)	32.2 (11%)	18.5 (5%)	11.2 (5%)	39.9 (11%)
Fe	mg/kg	2940(10%)	1.1% (10%)	2540(10%)	2670(10%)	1.1(10%)	6530 (10%)	1110 (10%)	539(10%)	400 (10%)	338(10%)	128(10%)	606 (10%)	932(10%)	833(10%)	532(10%)
Co	mg/kg	0.848 (8%)	2.11 (8%)	0.586 (8%)	0.52 (8%)	2.28 (8%)	1.4 (8%)	0.502 (8%)	0.182(8%)	6.05 (8%)	0.163 (8%)	0.0817 (8%)	0.349 (8%)	0.232 (8%)	0.209 (8%)	0.295(8%)
Zn	mg/kg	12.8 (10%)	21.6 (10%)	9.99 (10%)	21.6 (10%)	30.4 (10%)	26.2 (10%)	11.2 (10%)	4.83(10%)	27.2 (10%)	8.9 (10%)	2.87 (10%)	2.12 (10%)	9.02 (10%)	4.13 (10%)	29.9(10%)
As	mg/kg	0.82 (5%)	0.37 (5%)	0.304 (5%)	0.283 (5%)	1.08 (5%)	1.47 (5%)	2.39 (5%)	0.541(5%)	17.6 (5%)	0.782 (5%)	0.139 (5%)	0.852 (5%)	0.558 (5%)	0.334 (5%)	9.96 (5%)
Br	mg/kg	22 (3%)	26 (3%)	5.53 (3%)	11.6 (3%)	38.4 (3%)	39.8 (3%)	95 (3%)	51.1(3%)	599 (3%)	13.7 (3%)	5.42(3%)	11.3 (3%)	9.25 (3%)	4.32 (3%)	147(3%)
Rb	mg/kg	0.724(20%)	0.379(20%)	0.107(20%)	0.369(20%)	1.37 (20%)	1.81 (20%)	1.31 (20%)	0.558(20%)	3.18 (20%)	0.469(20%)	0.195 (20%)	0.612 (20%)	0.476(20%)	0.40 (20%)	1.32(20%)
Sr	mg/kg	5840 (7%)	4820 (7%)	3410 (7%)	5870 (7%)	4620(7%)	300 (7%)	4970(7%)	5490(7%)	2290(7%)	5130(7%)	5530(7%)	472(7%)	6220(7%)	5070(7%)	2360(7%)
Sb	mg/kg	0.033(15%)	0.031(15%)	0.018 (15%)	0.017 (15%)	0.089 (15%)	0.114(15%)	0.059(15%)	0.031(15%)	0.115(15%)	0.034(15%)	0.022(15%)	0.025 (15%)	0.042(15%)	0.035(15%)	0.046(15%)
I	mg/kg	8.45 (15%)	5.95 (15%)	2.04 (15%)	1.97 (15%)	23.9 (15%)	35 (15%)	9.44 (15%)	17.9 (15%)	42.3 (15%)	3.92 (15%)	2.78 (15%)	14.8 (15%)	2.66 (15%)	3.81 (15%)	8.34 (15%)
Ba	mg/kg	195 (10%)	40.3 (10%)	13.4 (10%)	12.1 (10%)	15.8 (10%)	18.9 (10%)	27.6 (10%)	14.5 (10%)	18 (10%)	9.94 (10%)	10.2 (10%)	2.87 (25%)	10.1 (10%)	8.25 (10%)	6.42 (13%)

Cs	mg/kg	0.03(15%)	0.0716(15%)	0.038 (15%)	0.0197(15%)	0.0638(15%)	0.186(15%)	0.0401(15%)	0.0253(15%)	0.083(15%)	0.032(15%)	0.01(15%)	0.018 (15%)	0.023 (15%)	0.021 (15%)	0.059 (15%)
La	mg/kg	1.57 (6%)	2.35 (6%)	0.621 (6%)	1.74 (6%)	8.12 (6%)	4.92 (6%)	2.6 (6%)	0.545 (6%)	<1.5	2.35 (6%)	0.531 (6%)	0.299 (10%)	0.315 (6%)	0.452 (6%)	0.56 (30%)
Ce	mg/kg	2.21 (25%)	<1.5	1.63 (16%)	4.08 (8%)	16 (7%)	11.6 (7%)	5.9 (11%)	1.72 (25%)	0.945 (25%)	5.92 (9%)	<1.5	<2.5	<1.5	0.747(12%)	2.23(30%)
Gd	mg/kg	0.35 (8%)	1.65 (8%)	0.352 (8%)	0.694 (8%)	2.5 (8%)	1.55 (8%)	0.695 (8%)	0.396 (8%)	0.136 (8%)	0.068(8%)	0.056(8%)	0.106 (8%)	0.619(8%)	0.619(8%)	0.196(8%)
Tb	mg/kg	0.0877 (6%)	0.303 (6%)	0.066 (6%)	0.080 (6%)	0.336 (6%)	0.196 (6%)	0.05 (6%)	0.0236 (6%)	0.016 (6%)	0.030 (6%)	0.009 (6%)	0.0178 (13%)	0.0211 (6%)	0.0172 (6%)	0.010 (11%)
Hf	mg/kg	0.427 (30%)	0.553 (30%)	0.094 (30%)	0.261 (30%)	2.59 (30%)	2.99 (30%)	3.63 (30%)	0.675 (30%)	0.326 (30%)	0.61 (30%)	1.03 (30%)	0.088 (30%)	0.22 (30%)	0.458 (30%)	0.136(30%)
Ta	mg/kg	0.044 (6%)	0.183 (6%)	0.021 (6%)	0.049 (6%)	0.175 (6%)	0.166 (2.7%)	0.071 (6%)	0.020 (6%)	0.009 (8%)	0.014(6%)	0.0075 (6%)	0.018 (25%)	0.0176 (6%)	0.018 (6%)	0.022(8%)
Th	mg/kg	1.07 (6%)	0.922 (6%)	0.37 (6%)	1.57 (6%)	4.63 (6%)	2.88 (6%)	1 (6%)	0.0197 (6%)	0.080 (20%)	1.46 (6%)	0.47 (6%)	0.090 (10%)	0.03 (20%)	0.018 (6%)	0.028(25%)
U	mg/kg	2.78 (6%)	2.75 (6%)	1.53 (6%)	2.68 (6%)	2.68 (6%)	1.46 (6%)	2.26 (6%)	2.95 (6%)	1.22 (6%)	2.03 (6%)	2.28 (6%)	0.222 (6%)	2.7 (6%)	2.16 (6%)	0.95(6%)



Discussion:

Corals ingest follow components into their skeletons from the encompassing water in which they develop. the lattice bound trace metals in coral skeletal structure reflect their concentrations in surface water where the corals grew (Inoue et al. 2004) Therefore, the trace element levels in coral skeletons may function as good proxies for marine pollution.(S. Krishna Kumar,2010). Mostly corals grow in a pristine environment, but are affected by near shore developmental activities such as coastal mining, harbour dredging, discharge of industrial and domestic effluents into the ocean, urbanization and over population (AnuGopinath et al. 2009). There are many potential sources which contribute in the pollution of Red sea. Therefore, this article presented most the potential sources and activities in Suez Gulf. The main cause appears is shipment of mineral items ,oil filling and seaside development, transport based sewage and solid waste, anti-fouling paints, land traffic, agriculture activities, and constructions.Chart Fig. (6) Expresses the concentrations of heavy metals (Ti, V, Mn,Fe,Zn, As and Sb) among the different species in the three phases, (Raw,clean and precipitate). As we see from charts Iron has the highest value of metal concentration in all phases samples (Raw, clean and precipitate) that indicates it is not only in the crust of the coral but composed in coral during its life time years ago. Also this means that coral is good archive for historical pollution of human activities. Hence trace metals can be incorporated into corals during their lives by a varied of mechanisms: substitution of dissolved metals species into crystal lattice (e.g. substitution for Ca), trapping of particulate (detritus) matter within skeletal cavities, uptake of organic matter from coral tissue, and coral feeding (Brown 1987; Hanna and Muir 1990; Ferrier-Pagès et al. 2005). Iron concentrations also is much higher than those concentration values of Iron in recent coral taken from (El Hamarwein, Sharm Al Bahari and Shalateen) in the Red sea of Egypt (Abdelbaset S. El-Sorogy,2012) , and coral samples taken from Aqaba Gulf of Jourdan in the Red sea (Al-Rousan,2007). Zn is essential elements for living organisms and plays an important role in growth, cell metabolism and survival of most animals including corals. Moreover, some microorganisms often tolerate greater concentration of certain metal compounds than can higher forms of life; here Zn is almost in the same average value with the mean value of Zn in recent coral in other mentioned Egyptian sites along Red sea (Abdelbaset S. El-Sorogy, 2012). Mn concentration in the corals is an indicator of detrital inputs (Linn et al. 1990), which is also, has higher value in comparison with other sites in the Red sea of Egypt. The most likely mechanism for incorporation of metals in corals is a combination of uptake of seawater-soluble metals and incorporation via polyp feeding (Fallon et al. 2002; Ramos et al. 2004).Even though

Without knowledge of the fractionation factors involved and their causes, corals cannot be used to determine the trace element composition of adjacent waters (H. Herbert Veeh and Karl K. Turekian, 1968). Ba founded in all species and it could be an indicator of contamination by land use (Prouty et al. 2010) and flood events (Sinclair, 2005).

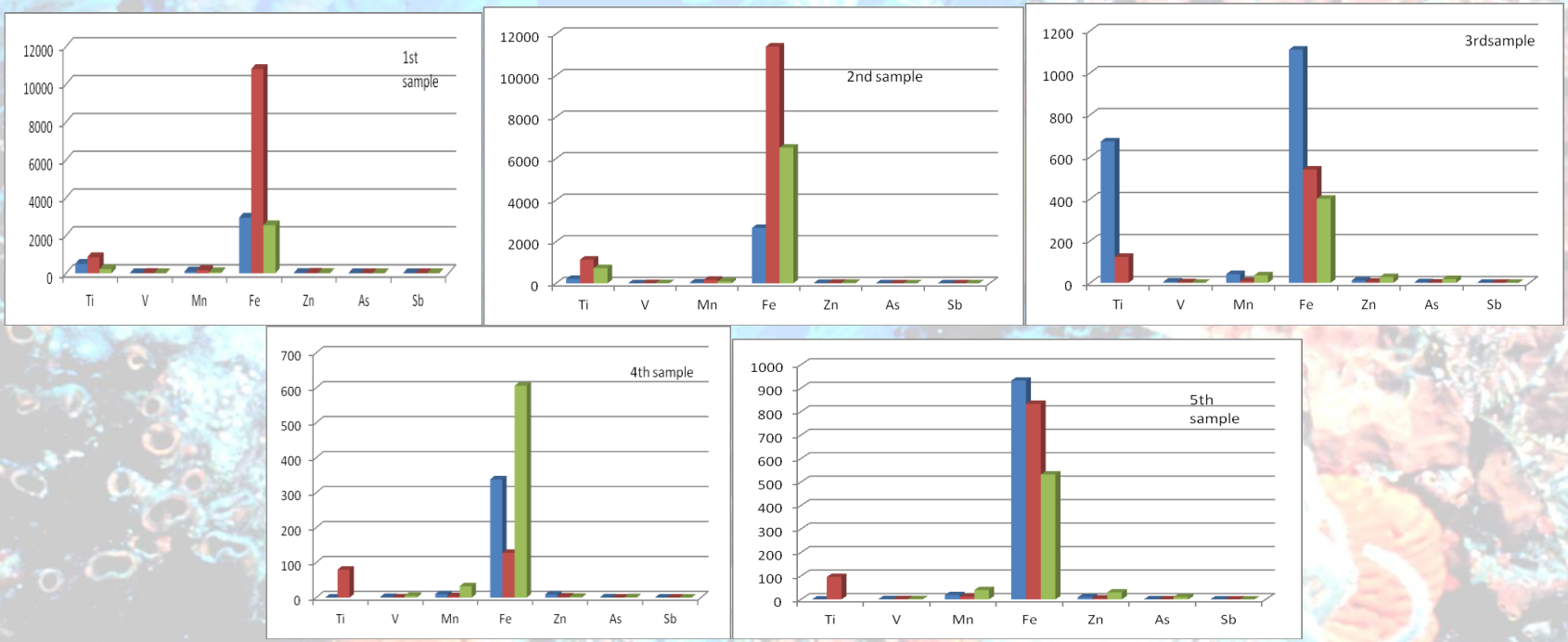


Figure 3. Concentrations of elements in (Raw, Clean and precipitate)

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