International Journal of Advanced Biotechnology and Research (IJBR)
ISSN 0976-2612, Online ISSN 2278–599X,
Vol-9, Issue-2, 2018, pp418-427
http://www.bipublication.com

Research Article

A Novel Cluster Analysis of Scrutiny Anzali Wetland Pollution to Heavy Metals with Data Mining Techniques

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ABSTRACT:

Scrutiny of heavy metals as toxic pollutants in environment has a significant importance in environmental pollution studies. surficial sediments of water resources have a high potential in releasing heavy metals to the upper water environment, hence sediment analysis presents guidelines to the authorities for monitoring the environmental systems. In this study, total concentration of Five heavy metals (Ni, Zn, As, Fe, and Pb) were investigated along different sites of Anzali Wetland in Iran using cluster analysis and bulk elemental analysis behind data mining techniques and chemical extraction techniques respectively. Geoaccumulation index (Igeo) and Pollution Index (Ipoll) were computed and compared in different sites of the wetland, too. Total concentration of metals in sediment samples found to be in this Research: Fe>As> Zn>Ni> Pb. The speciation data revealed that most metals were bonded in lithogenous fractions that means no pollution. Igeo results indicated that the wetland is moderately to highly polluted for As and Ipoll results showed that the wetland is moderately polluted for Pb were applied to metals also to investigate on Igeo and Ipoll results. For Ni, Zn and Fe, cluster analysis confirmed both indices but for As and Pb it confirmed Igeo results.The results of the present study showed that the Anzali wetland is threatened by pollutants related to rivers entering it. So to preserve the environment of the Anzali, the main act is to prevent the discharge of wastewater to rivers entering it.

Keywords: Anzali Wetland, Heavy metals, Cluster Analysis, Geoaccumulation Index (I_{geo}), Pollution Index (I_{poll}), Data mining.

[I] NTRODUCTION

Lokeshwari and Chandrappa assessed the level of some heavy metals in water, water hyacinth and sediment samples of Lalbagh tank, Bangalore, India [6].

Geoaccumulation index results revealed that there was moderate input of copper and lead from anthropogenic sources to the tank basin. Priju and Narayana analyzed five heavy metals on sediments of Vembanad Lake in India to understand the pollution levels and the impact on the coastal environment [7]. They showed that industrial effluents are major source of metal enrichment in the lagoonal system. Praveena, et al. tried to establish a complete and comprehensive set of sediment quality guidelines by applying numerous sediment quality guidelines on Mengkabong lagoon mangrove sediment in Malaysia [8]. They concluded that the most appropriate guideline that meets the prioritization criteria consistent with international initiatives and regulations is interim sediment quality values for Hong Kong. In another research, Praveena, et al. investigated on Mengkabong lagoon mangrove sediments by applying principal components analysis (PCA) and cluster analysis (CA) to interpret information about the sediment and its controlling factors [9]. Calculation of geoaccumulation index indicated that the Mengkabong mangrove sediments are having background concentrations for Al, Cu, Fe, and Zn and unpolluted for Pb. Lasheen and Ammar investigated the mobility and the availability of eight heavy metals in sediments from different sites along the Nile River in Cairo district using sequential chemical extraction technique to evaluate their threats to ecological and human health [10]. Banat and Howari evaluated the accumulation of six heavy metals in fine-grained sediments from the sediments of the late marshes and swamps of southern Iraq [11]. They reported that the concentrations of Co, Zn, Cr and Cu approach the average concentration in the Earth's crust and uncontaminated sediments but Pb, Ni, and Cd are slightly enriched which may reflect anthropogenic effect. It was believed that metals in the study area were derived mainly from the igneous mineral deposits in the IraqIran Mountain range. Mohiuddin, et al. investigated seasonal and spatial distribution of heavy metals in the bed sediments of polluted Tsurumi River in Japan by analyzing ten heavy metals and revealed that Tsurumi River sediments are moderately to heavily contaminate by Zn, Pb and Co base on Pollution Index (I_{poll}) [12].

Nasrabadi, et al. measured total content and four chemical partitioning fractions of ten heavy metals in Haraz River bed sediments in Iran and concluded about its contamination level [13]. Yu, et al. investigated the effect of Mo mining activities in western Liaoning, northeast China with geochemical method and showed that Mo tailings ponds deposited along the bank may have a closely relationship with the high levels of these metals in sediments and Mo in sediments may pose a high risk to the local environment [14]. Hosseini, et al. determined the concentration of some elements in water and sediment of Shadegan wetland, Iran [15]. They measured I_{geo} and I_{poll} Indices and discussed about the wetland metal contamination base on the Indices.

The Anzali Wetland, located on the southern coast of the Caspian Sea in Gilan Province of Iran, is a large complex of freshwater lagoons with extensive reed-beds, shallow impoundments and seasonal flooded meadows. It is internationally known as an important wetland for migratory birds and was registered as a Ramsar site in 1975. The water quality of the wetland is deteriorating due to the inflow of domestic, agricultural and industrial wastewater from neighboring cities. A few researches on evaluation of metal pollutions have been done on Anzali wetland. Pourang measured Lead, Copper, Zinc and Manganese in surfacial sediments of the wetland [16]. Results indicated that no distinct relationship existed between heavy metal levels and percentage fine fraction in sediments. Amini-Ranjbar determined the concentrations of Cd, Pb, Cu, Zn and Ni in surficial sediments from eleven sampling sites in the Anzali wetland and statistically proved significant differences among the accumulation of the metals in sediments, while differences were not observed among the seasons [17]. To investigate the precipitation of heavy metals in Anzali wetland and evaluate its refining performance, Sartaj, et al. collected sediment samples from ten stations including inlets, outlets and some internal locations in the wetland and analyzed them for five metals of As, Fe, Pb, Zn, Cu and Ni over a period of six months [18]. Their results indicated that concentration of heavy metals decreases with an increase in the distance from delta of rivers entering the wetland. This is due to the role and performance of wetland chemical contents in reducing the pollutants, the self-purification action of wetland as well as precipitation of heavy metals at the beginning of the entries into the wetland. Significant differences between the sampling sites in Anzali wetland from the concentrations of cadmium, copper, and lead in surficial sediment were reported by Pourang, Richardson and Mortazavi [19]. The mean concentrations of Pb in the sediments were higher than the global baseline values and world average shale. Ghazban and Zare investigated on the pollution situation of Anzali wetland by analyzing heavy metals in core sediments of the wetland [20]. They measured major elements and trace elements of sediment samples and compared their results with the Caspian Sea and global sediments and showed that the concentrations of heavy metals in wetland sediments are higher.

In this study sediment samples were collected from ten stations along the Anzali wetland and the concentration of five heavy metals Ni, Zn, As, Fe and Pb were measured by total elemental analysis method [21]. Tessier Chemical partitioning method was applied to recognize the kind of heavy metal bonds to sediment and to know the background concentration of metals in sediment [22]. Geoaccumulation index (I_{geo}) and pollution Index (I_{poll}) were measured and compared with each other. Cluster Analysis was applied to investigate the relationship between different metals and to assess the I_{geo} and I_{poll} results.

[II]STUDY AREA AND SAMPLING SITES

The Anzali wetland is a large complex environment of fresh water lagoons with extensive reed-beds, shallow impoundments and seasonal flooded meadows. . It is wetland located in the northern part of Iran, along the coast of the Caspian Sea approximately at north latitude between 37o 25′ and 37o 32′ and east longitude between 49o 15′ and 49 o 36′. It has a catchment area of 3610 km2. Approximately 41.5% of the catchment area is covered by forests. Among the landuse categories, forest has the largest share of 41.5%, followed by paddy field/farmland (26.9%) and orchard (8.7%) in that order Figure 1. There are ten major river systems entering the wetland. The annual mean discharge into the wetland is estimated at 76.14 m3/s. Base on inflowing rivers the wetland can be divided as two zones: the west zone that has only one river inflowing (Zone A) and the southern and eastern zone that has nine rivers inflowing (Zone B) Figure 2. The average annual of Anzali Wetland watershed is about 1200 mm. Ten sampling sites were chosen along the wetland considering the situation of the wetland (low depth marsh areas) and sediment samples were collected. Figure 2 shows sampling sites. Samples were collected using Ekman grab sampler on May 2014. Samples were dried in an oven at 105oC and powdered in an agate mortar. In order to normalize the variations in grain size distributions, the dried sediment samples were sieved to 0.15 mm using sieve No. 150.

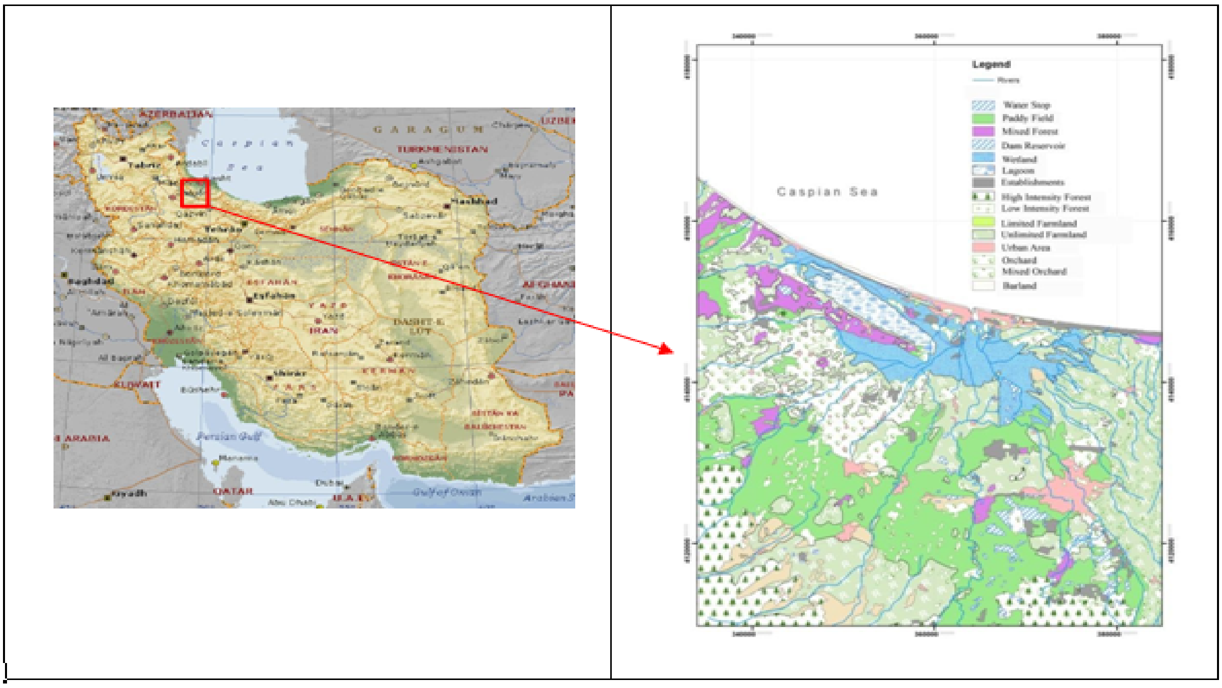

Fig: 1. Anzali wetland location and landuse of its basin

[III] MATERIALS AND METHODS

3.1. Total elemental analysis

Digestion of organic matter and dissolution of silicates for total elemental analysis was proposed by Sparks, et al. [21]. The procedure is described below: 1.0 g of the 100-mesh (0.15mm) sediment was weighed into a 100-mL Teflon beaker and 10 ml of HNO3 and 10 ml of HClO4 were added. The beaker was covered with a Teflon watch cover and heated at 200oC for one hour. The cover was removed and heating was continued until the volume became 2 to 3 ml. After cooling the sample, 5 ml of HClO4 and 10 ml of HF were added; Teflon cover put and heated at 200oC until all siliceous materials has been dissolved. Then the cover removed and heating continued until the volume was 2 to 3 ml. The digest cooled, 10 ml of 50% HCl added, Teflon cover put and heated at 100oC for 30 minutes. After cooling the sample brought to 50-mL volume. The solution is then ready for ICP determination [21]. The concentrations of heavy metals were determined according to APHA (1998) [23]. The analyses of total digestion samples were duplicated.

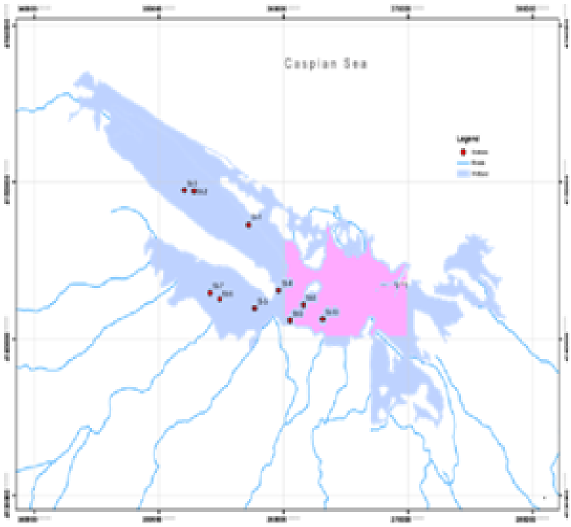

Fig 2. Sampling sites.

3. 2. Metals speciation

The metals speciation using sequential extraction proposed by Tessier, Campbell and Bission and modified by Elsokkary and Müller [22,24]. Heavy metals have five kinds of bonds so following five operational steps were used for partitioning the heavy metals.

3.2.1. First Stage (exchangeable)

One gram solid (sediment or sludge) sample was shaken for one hour at room temperature with 8 ml of 1 M magnesium chloride -6 hydrate (MgCl2.6H2O).

3.2.2. Second stage (bound to carbonates)

The residual solid from exchangeable fraction of metals was shaken for 30 minutes at room temperature with 8 ml of 1 M sodium acetate anhydrous ($C_2H_3NaO_2$) and adjusted to pH 5.0 with acetic acid (99.83% $C_2H_4O_2$).

3.2.3. Three stage (bound to iron and manganese oxides)

The residual solid from carbonate fraction of metals was shaken at 85oC in water bath for five hours with 20 ml of 0.04 M Hydroxylamine hydrochloride ($H_3NO.HCl$) in 25% acetic acid (99.83% $C_2H_4O_2$) (v/v).

3.2.4. Four stage (bound to organic and sulfide)

The residual solid from Fe/Mn-Oxide fraction of metals was shaken at 85oC in a water bath for two hours with 5 ml of hydrogen peroxide (30% H_2O_2). Nitric acid (0.02 M HNO_3) was added to reach the pH of samples to 2 ± 0.2 pH units. Subsequently, a second addition of 3 ml of 30% H_2O_2 was added, and pH was monitored during the experiments. Additional acid was added as necessary to maintain the pH of the samples within 2 ± 0.2 pH units and shaken again at 85oC in a water bath for three hours.

3.2.5. Five stage (residual)

Finally, the residual solid from the organic and sulfide fraction of metals was digested with a mixture of HNO_3, HF, $HClO_4$, HC_l in [4:1:1:1] ratio, respectively, for three hours in a water bath. The residue dissolved entirely, and it is diluted with 50 ml distilled water.

In each operational fraction step the extractions were conducted in centrifuge tubes (50 ml with cap) to minimize losses of solid materials, and centrifuged at 2000 rpm for 30 minutes. The supernatant was filtered through Whatman No. 4 filter paper, and the residue was washed, shaken with 8 ml of deionized water for 30 minutes, and centrifuged, so it was ready for the next step and the washing was combined with the supernatant of each step [25,26,10]. The concentrations of heavy metals in all steps were determined by Inductively Coupled Plasma Mass Spectrometry (ICP-MS) (Varian 710) according to APHA (1998). The analyses of the sequential extractions procedure were being replicated two times.

Table 1. Total concentration of heavy metals in sediments of the Anzali wetland and mean crust

Sites	Fe ppt	Zn ppm	As ppm	Pb ppm	Ni ppm
St1	22.80	106.50	17.50	18.31	96.75
St2	22.50	103.22	17.32	19.50	89.40
St3	21.25	106.51	18.60	18.82	94.50
St4	23.25	126.25	32.75	23.45	108.52
St5	23.20	117.50	31.67	27.25	112.12
St6	25.40	120.82	31.70	27.99	106.10
St7	26.58	122.65	35.45	27.70	114.01
St8	23.12	114.22	39.85	26.40	101.05
St9	23.62	123.40	29.10	26.42	108.00
St10	25.25	144.20	31.50	25.32	104.01
Min	20.30	103.22	17.30	18.30	86.40
Max	27.81	145.61	38.85	29.50	114.00
mean	23.87	120.98	28.74	24.65	103.52
Mean Crust	51.00	70.00	5.00	13.00	80.00

3.3. Geochemical Indices

To quantify the degree of anthropogenic contamination of heavy metals in different ranges of concentration in wetland sediments, index of geoacumulation, Igeo and index of pollution, Ipoll were used. The index of geoacumulation (Igeo) has been used as a measure of bottom sediment contamination since the 1970s, and numerous researchers have employed it to assess the contamination of soils and sediments. It determines contamination by comparing current metal contents with pre-industrial levels. The content accepted as background is multiplied each time by the constant 1.5 in order to take into account natural fluctuations of a given substance in the environment as well as very small anthropogenic influences. The value of the geoaccumulation index is described by the following equation [27]:

$$I_{geo} = Log_2\left[\frac{C_n}{1.5\times B_n}\right]$$

Where Cn is measured concentration in sediment mg/kg, Bn is geochemical background value mg/kg and the factor 1.5 is introduced to include possible variations of the background values that are due to lithologic variations. The interpretation of the obtained results is as follows: Igeo ≤ 0 practically uncontaminated, 0 < Igeo < 1 uncontaminated to moderately contaminated, 1 < Igeo < 2 moderately.

$$I_{Poll} = Log_2\left[\frac{B_c}{L_p}\right]$$

where Bc and Lp are indicative of, bulk concentration and lithogenous portion, respectively. Since there was not any need in these evaluations to use the shale metal concentrations, the constant factor (1.5) had been eliminated. Here, chemical partitioning results is substituted for the mean crust and shale levels. The interpretation of the obtained results is like Igeo.

In the present study Igeo and Ipoll were adopted to assess the pollution intensity in the wetland sediments.

[IV] RESULTS

Table 1 shows the results of total concentration of heavy metals in sediment samples and in the earth crust. As Table 1 indicates, mean concentration of elements in sediment samples follows this pattern: Fe>Zn>Ni>As>Pb. The concentration of Fe varies from 20.30 ppt in St3 to 27.81 ppt in St11. Zn varies from 103.20 ppm in St2 to 145.60 ppm in St10. As ranges between 17.30 ppm in St2 to 38.85 ppm in St8. Pb varies from 17.30 ppm in St2 to 29.5 ppm in St11. Ni varies between 86.40 ppm in St2 to 114.00 ppm in St7. It can be observed that the concentration of metals in the zone A of wetland (St1 to St3) is lower than the concentration in the zone B (St4 to St12. This contaminated, 2 < Igeo < 3 moderately to heavily contaminated, 3 < Igeo < 4 heavily contaminated, 4 < Igeo < 5 heavily to very heavily contaminated and Igeo ≥ 5 very heavily contaminated. Ipoll was presented as a new developed pollution index by karbassi (2008) to evaluate the pollution intensity of metals in sediments as follows: [2] concept is related to the rivers inflowing to the wetland. Industrial wastewater from Rasht industrial city is discharged to rivers and they inflow to the east zone of wetland Anzali as illustrated in Figure 2. Considering formula 1 and replacing data on Table 1 as Cn and the crust content of elements as Bn gives the value of Igeo for metals in each station. The results have been illustrated in Figure 3. Base on Igeo classification, As places in class 3 i.e. moderately to heavily contaminated. All the Heavy metals studied are located in class 1 and 0 that indicates no pollution.The result data from chemical partitioning on sediment samples are given in Table 2. Because of high experimental costs two stations from zone A (St1 and St3) and four stations from zone B (St5, St7, St9 and St11) were chosen for chemical partitioning.

Table 2. Specification of heavy metals in Anzali wetland sediments

Fractional Steps								
metals	**Stations**	**step1**	**step2**	**step3**	**step4**	**step5**	**nonlithogenous (%)**	**Lithogenous (%)**
Fe (ppt)	**St1**	0.25	0.65	4.3	3.5	14.5	12.4	87.6
	St3	0.20	0.60	4.10	3.50	15.30	10.7	89.3
	St5	1.10	3.10	4.80	5.90	17.60	17.7	82.3
	St7	1.00	3.10	4.50	5.20	16.50	18.4	81.6
	St9	1.20	3.20	5.60	5.10	16.20	21.9	78.1
	Mean	0.81	2.19	4.75	4.80	15.98	16.71	83.29

metals	Stations	step1	step2	step3	step4	step5	nonlithogenous (%)	Lithogenous (%)
Zn (ppm)	**St1**	11.30	18.60	13.20	23.00	44.20	29.1 28.6	70.9
	St3	12.30	16.30	14.50	22.30	46.20	35.4	71.4
	St5	18.60	23.50	16.80	23.50	47.20	35.0	64.6
	St7	21.00	23.50	16.30	24.60	49.60		65.0
	St9	22.30	25.60	17.50	24.20	48.30	38.9	61.1
	Mean	17.80	21.77	16.32	23.47	46.62	34.05	65.95

metals	Stations	step1	step2	step3	step4	step5	nonlithogenous (%)	Lithogenous (%)
As (ppm)	**St1**	1.50	4.10	5.10	3.20	10.10	34.6	65.4
	St3	1.90	3.80	5.20	3.60	10.50	33.6	66.4
	St5	4.20	6.80	6.80	4.80	11.30	42.5	57.5
	St7	5.60	6.30	6.50	5.00	12.30	41.5	58.5
	St9	5.30	7.50	6.40	4.90	11.60	43.8	56.2
	Mean	4.02	5.87	5.98	4.37	11.13	39.95	60.05

metals	Stations	step1	step2	step3	step4	step5	nonlithogenous (%)	Lithogenous (%)
Pb (ppm)	**St1**	2.30	5.30	1.30	3.60	6.70	36.4	63.6
	St3	3.10	5.70	1.50	3.80	7.50	37.7	62.3
	St5	4.80	8.30	1.80	3.50	6.80	49.1	50.9
	St7	5.30	8.50	1.60	3.20	6.90	50.4	49.6
	St9	5.60	8.60	2.20	3.10	5.20	56.4	43.6
	Mean	4.80	8.08	2.04	3.36	7.91	49.90	50.10

metals	Stations	step1	step2	step3	step4	step5	nonlithogenous (%)	Lithogenous (%)
Ni (ppm)	**St1**	7.10	2.60	11.10	34.50	49.30	9.9	90.1
	St3	8.40	2.50	12.30	34.30	46.00	11.3 17.5	88.7
	St5	12.60	5.20	15.60	35.00	51.20	16.7	82.5
	St7	12.30	5.30	15.20	35.20	53.00		83.3
	St9	13.50	5.60	14.80	32.10	54.80	20.2	79.8
	Mean	11.82	5.08	14.94	33.96	50.74	17.26	82.74

Step 1 and step 2 (exchangeable and carbonate forms) are the easily assimilable fractions and shows the greatest degree of metal mobility. Step 3 and step 4 (Fe/Mn-oxide and organic and sulfide forms) exhibit some degree of mobility, and the step 5 (residual form) corresponds to the part of the metals which cannot be mobilized.

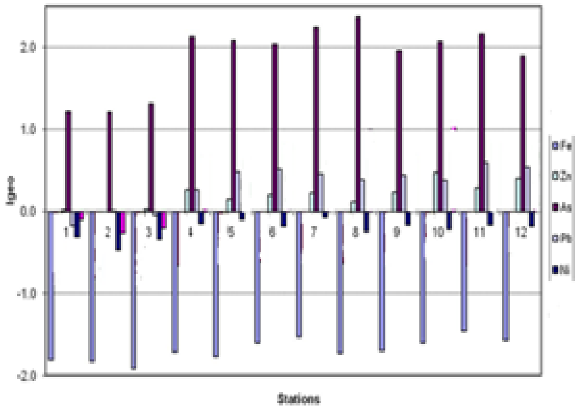

Fig3. Igeo of metal in Different sites.

Table 2 indicates that the mean value of lithogenous part of metals in sediments follows this pattern: Fe(83.29%) > Ni[82.74% > Z n(65.95%) >As (60.05%) > Pb(50.10 . Figure 4 compares the lithogenous and anthropogenic (nonlithogenic) percentage of metals in selected stations. It is obvious that the lithogenous part of stations 1 and 3 (zone A) is higher than other stations (zone B).

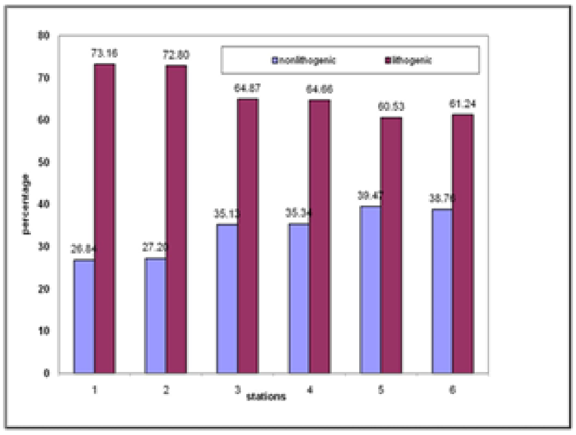

Fig 4. Comparison of nonlithogentic and lithogentic part of heavy metal in Anzali weland

This fact can be observed also by computing Ipoll. Considering formula 2, and replacing data on Table 1 as Bc and lithogenous data from Table 2 as Lp gives the value of Ipoll for metals in selected stations. Figure 5 illustrates the results. Lp is calculated in this way: (step4+step5)+0.1(total content)[5].

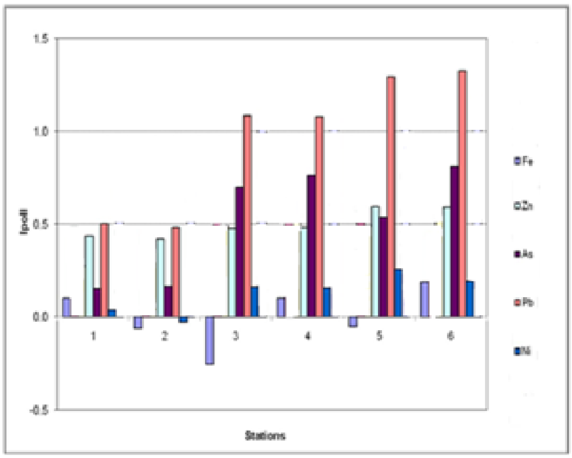

Fig 5. Ipoll of metals in stations

Figure 5 shows that base on Ipoll, only Pb in zone B of the wetland are in class 2 i.e. moderately contaminated and most of metals categorize in class 1 i.e. no pollution. Arsenic is in class 3 even though base on Ipoll it is in class 1 and for Pb, it is in class 1 in Igeo but in class 2 base on Ipoll.The difference between these indices for selected stations is illustrated in Figure 6. To investigate on this difference the cluster analysis was run on the metals base on data from Table 1.

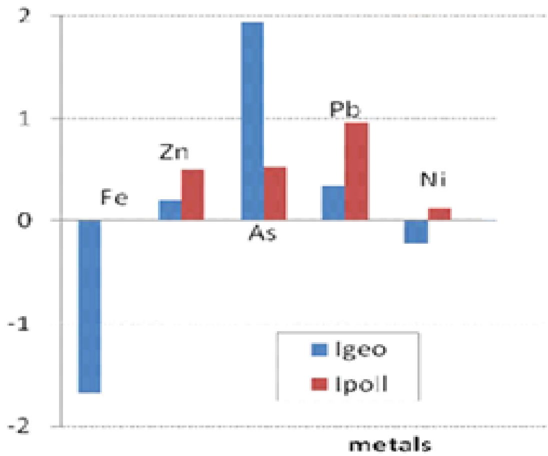

Fig 6. Comparison between mean I_{geo} and mean I_{poll}

Results have shown in Figure 7. The cluster analysis indicated that there is strong relationship amongst all metals (Pearson coefficient> 0.75) especially between Pb and Fe.

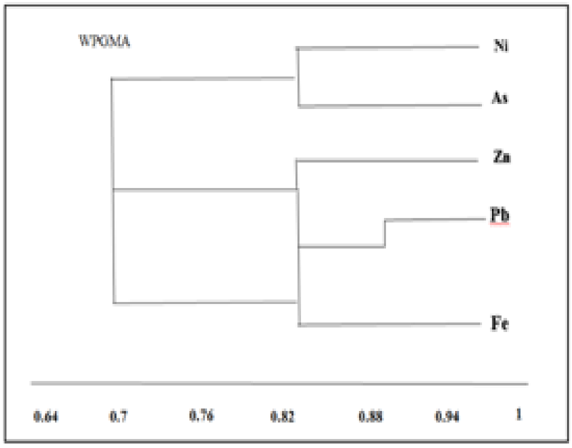

Fig 7. cluster analysis of heavy metals in Anzali wetland

Because Fe originates from natural sources it can be resulted that Pb originate from natural sources, too [5]. It means cluster analysis confirms Igeo for Pb. As has strong relationship with Ni. Since Ni originates from anthropogenic sources As comes from anthropogenic sources, too [5]. It means cluster analysis verifies Igeo results. Considering this fact that the discharge of industrial wastewater to the Anzali wetland from input rivers is the most important reason for its contamination, it can be concluded that in the case of Anzali wetland, Igeo is more reliable than Ipoll.

[V] CONCLUSION

Pb but to achieve the contamination level of The main objective of this study was to wetland, Igeo and Ipoll as two pollutin indices were computed and compared together. The evaluate the contamination of Anzali wetland to heavy metals. To achieve the aim, ten results showed base on Igeo, As stations along the wetland were chosen and base on Ipoll Pb are pollutants for the wetland. The results of this study showed that surfacial sediment samples were collected.Five heavy metals Ni, Zn, As, Fe, Ipoll index can be effectively applied to show and Pb were analyzed by total elemental environmental pollution like Igeo because it uses background concentrations of metals base analysis and sequential chemical extraction on the status of the study area but it is better to techniques. Comparing the results of measuring check the results by statistical analysis. In this the concentration of mentioned metals with the study Cluster analysis was applied. It mean concentration of earth crust showed high contamination level for Zn, As, Ni and confirmed the results of Igeo and Ipoll for Ni, Fe, Zn but for As and Pb it confirmed Igeo results. Totally, the results of the present study showed that the Anzali wetland is threatened by pollutants related to rivers entering it because the west zone of wetland that only one river inflows (zone A), the pollution indices have lower magnitude in contrast to the other zone of wetland that nine rivers inflow (zone B). So to preserve the environment of the Anzali wetland from deterioration, the main act is to prevent the discharge of wastewater to rivers entering it.

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