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Calculating Integrated Pollution Indices for Heavy Metals in Ecological Geochemistry Assessment Near Sugar Mill

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ABSTRACT:

The sugar mill is a good example of a site where human pressures and ecological values collide with each other. One of the aims of this work was to select different types of index to aggregate and assess heavy metal contamination near sugar mill in an accessible manner. Concentrations of heavy metals (Iron, Manganese, Zinc and Copper) are studied in the soil near sugar mill to asses metal contamination due to industrialization. The soil samples were collected from three different depths A (0 cm), B (5 cm) and C (10 cm) for a period between October 2010 and March 2011 (winter and summer) and the heavy metal contents were analyzed by Atomic Absorption Spectrophotometer. Pollution index is a powerful tool for ecological geochemistry assessment. Nine integrated indices were divided into two groups. One group is suitable for the normal distribution single indices including the average, vector modulus, and Nemerow pollution indices, and the other for log-normal distribution including the product, root of the product, and weighted power product pollution indices. Using background levels as reference values, five contamination classes were divided, and the terminologies are suggested for the integrated indices to unify the assessment results. The pollution load index (Ecological risk index) indicates that soil near sugar mill was highly polluted due to heavy metals (PLI_{Fe} = 0.30, PLI_{Mn} = 0.58, PLI_{Zn} = 0.24 and PLI_{Cu} = 0.34). The results of contamination index, index for chemistry and metal pollution were in agreement with pollution load index. Average and vector modulus of pollution index and Nemerow pollution index indicated slightly polluted domain. Since the aim of work on contamination evaluation is to assess the overall contamination of a study area, the indices are highly appropriate.

Keywords:

Atomic Absorption Spectrophotometer, integrated indices, pollution index, heavy metals, ecological risk index, Nemerow pollution index.

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INTRODUCTION

With increasing population and industrial expansion, the need for the treatment and disposal of the waste has grown (Sarala and Sabitha, 2012a; Sarala and Sabitha, 2012b). Pollution of the natural environment by heavy metals is a worldwide problem because these metals are indestructible and most of them have toxic effects on living organisms (Sarala and Sabitha, 2009a). There is increased awareness that heavy metals present in the soil may have negative consequences on human health and on the environment (Sarala *et al*., 2009b).

From the environmental point of view, all heavy metals are largely immobile in the soil system, so they tend to accumulate and persist in agricultural soils for a long time. The most frequently reported heavy metals with potential hazards in soils are Cadmium, Chromium, Lead, zinc and copper (Alloway, 1995). The concentration of these toxic elements in soils may increase from various sources including anthropogenic pollution, weathering of natural high background rocks and metal deposits (Senesi *et al*., 1999). These chemicals in the terrestrial environment clearly pose a significant risk to the quality of soils, plants, natural waters and human health (Hooda and Naidu., 2004; U.S.E.P.A, 2003; W.H.O, 2004; Sarala and Sabitha., 2011).

With the development of ecological geochemistry survey and exploration geochemistry survey, a great deal of data related to heavy metal concentration in soils and water sediments have been measured which can be used to assess the quality of ecological geochemistry environment. Many calculation methods have been presented to assess the environmental quality, such as pollution index, principle component analysis (Cheng *et al*., 2007), gray correlation, and fuzzy decision. Different calculation methods on the basis of different algorithms might lead to discrepancy on pollution assessment when they are used to assess the quality of soil and/or sediment ecological geochemistry. So it is of great importance to select a suitable method to assess soil and sediment quality for decision making and spatial planning.

Pollution index is a powerful tool for processing, analyzing, and conveying raw environmental information to decision makers, managers, technicians, and the public (Caeiro *et al*., 2005). This article presents the results from the study of pollution indices by heavy metals in ecological geochemistry assessment. The aim of this work is to select different types of indices to aggregate and assess the heavy metal contamination near sugar mill. The different types of indices are compared and discussed.

MATERIALS AND METHODS

In the present study, stratified regular sampling method was adopted for soil sample collection as in geo-assessment of the variables estimated; the stratified regular sampling is more suitable because this kind of sampling draws homogenous error (Burges and Webster, 1980; Burges *et al*., 1981). For this purpose the grid map of the study area has been used to know the distribution of heavy metal concentration in the whole region by stratifying the region into a regular-sized grid cells, each grid cell is further divided into many smaller subcells for a period between October 2010 and March 2011 (winter and summer). Five sampling points in a grid of 0.5×1km at each sampling station (4 at the corners and one at the centre of the grid) were selected and composite samples consisting of three sub-samples were collected from the top (0 to 10 cm) layer of the soil using plastic spatula after removing the debris, rock pieces and physical contaminants. In order to have the background concentration values of the heavy metal elements, three soil samples were collected, each from 100 cm below ground level, which are least affected by the sugar mill. The samples were placed in the clean polythene bags, which were brought to the laboratory.

Laboratory methodology

The samples were brought to the laboratory where they were air dried and mixed thoroughly to obtain the representative samples. Soon after drying the debris and other objects were hand picked up and the sample was ground in a mortar to break up the aggregates or lumps, taking care not to break actual soil particles. Soil samples were then passed through a 2mm sieve in order to collect granulometric fraction. Since trace metals are often found mainly in clay and silt fractions of soil and hence the size fraction <63µm is most commonly in the recommended size. For this purpose the granulometric fraction was added with the dispersing agent and after shaking the sand fraction was separated from the clay and silt with <63µm sieve (wet sieving) and was used to measure the concentration of the heavy metals Fe, Mn, Zn and Cu from all the samples collected.

For this purpose the clay and silt fraction were digested by acids to get the solution by taking 5g of sample into a 300ml polypropylene wide-mouthed jar and distilled water was added to make a total 200ml. Then it was acidified with 10ml HF, 5ml $HClO_4$, 2.5ml HCl and 2.5ml HNO_3 in order to completely digest the soil. This jar was shaken on an orbital shaker for 16 hours at 200-220 rpm before being filtered through whatman filter paper (No. 42) into acid washed bottles. The solution was stored and heavy metal contents were analyzed by Atomic Absorption Spectrophotometer as per the method recommended by Committee of Soil Standard Methods for Analyses and Measurement (1986).

Pollution Indices

Caeiro *et al.*, (2005) analyzed the pollution indices to assess heavy metal contamination and classified them into two types: (i) contamination indices and (ii) ecological risk indices.

Integrated Indices

Integrated indices are indicators used to calculate more than one metal contamination, which were based on the single indices. Each kind of integrated index might be composed by the above single indices separately. According to algorithm, eight integrated methods were illustrated as following.

Average of Pollution Index

An average of pollution index (PI_{Avg}) can be defined as

$$PI_{Avg} = \frac{1}{m} \sum_{i=1}^{m} P_i$$

where Pi is the single pollution index of heavy metal i, and m is the count of the heavy metal species. This kind of pollution index was used by Bhattacharya *et al.*, (2006). A PI_{Avg} value of >1.0 indicates low quality soil because of contamination.

Vector Modulus of Pollution Index

A vector modulus of pollution index ($PI_{vectorM}$) can be defined as

$$PI_{vectorM} = \left(\frac{1}{m} \sum_{i=1}^{m} P_i^2 \right)^{1/2}$$

where Pi is the single pollution index of heavy metal i and m is the count of the heavy metal species.

Nemerow Pollution Index

A Nemerow pollution index ($PI_{Nemerow}$) was applied to assess the quality of soil environment widely (Cheng *et al.*, 2007) and was defined as

$$PI_{Nemerow} = \left(\frac{\left(\frac{1}{m} \sum_{i=1}^{m} P_i \right)^2 + P_{imax}^2}{2} \right)^{1/2}$$

where P_i is the single pollution index of heavy metal i; P_{imax} is the maximum value of the single pollution indices of all heavy metals, and m is the count of the heavy metal species. The quality of soil environment was classified into five grades from

Nemerow pollution index: $PI_{Nemerow} < 0.7$, safety domain; $0.7 \leq PI_{Nemerow} < 1.0$, precaution domain; $1.0 \leq PI_{Nemerow} < 2.0$, slightly polluted domain; $2.0 \leq PI_{Nemerow} < 3.0$, moderately polluted domain; and $PI_{Nemerow} > 3.0$, seriously polluted domain by Cheng *et al*., (2007).

Pollution Index

Johansson and Johnsson (1976) and Ott (1978) developed pollution index (contamination index) which was given by the formula

$$PI = \sum_{i=1}^{n} W_i C_i$$

where W_i is the weight for pollution variable i; C_i the highest concentration of pollution variable i reported in a location of interest. For each pollutant i, the weight was based on the reciprocal of the median of observed concentrations.

This index allows the identification of priority contaminations sites for implementation of decontamination action. It requires several measurements in the same sampling location. No threshold classification from unpolluted to high pollution.

Pollution Load Index

Wilson and Jeffrey (1987) framed the pollution load index (ecological risk index) as follows:

$$PLI = \text{antilog}_{10} \left(1 - \frac{C - B}{T - B} \right)$$

where B is the baseline value not contaminated; T the threshold, minimum concentrations associated with degradation or changes in the quality of the estuarine system. Wilson and Jeffrey (1987) defined B and T for the different contaminants; C the concentration of the pollutant. For each place the PLI calculation takes into account all the n contaminants:

$$PLI = (PLI_1, PLI_2, \ldots\ldots PLI_n)^{1/n}$$

Varies from 10 (unpolluted) to 0 (highly polluted).

Metal Pollution Index

Metal pollution index was given by Usero *et al*., (1996) which was categorized as contamination index.

$$MPI = (M_1, M_2, \ldots\ldots M_n)^{1/n}$$

where M_n is the concentration of metal n expressed in mg/kg of dry weight.

Metal Enrichment Index

Riba *et al*., (2002a) developed the metal enrichment index (contamination index) as follows:

$$SEF = \frac{C_i - C_o}{C_o}$$

where Ci is the total concentration of each metal i; C_0 the heavy metal background level established for the ecosystem studied.

Potential Ecological Risk Index

Potential ecological risk index (ecological risk index) was framed by Riba *et al*., (2002a) as given below:

$$ERF = \frac{C_i - C_{SQV}}{C_{SQV}}$$

where Ci is the total concentration of each metal i measured; C_{SQV} the highest concentration of the heavy metal non-associated with biological effects (chemical concentration associated with adverse effects); polluted stations have values equal to or greater than 1.

Five classes of terminologies were suitable to describe the degree of contamination. The five classes of contamination degrees, and their terminologies for soils were tabulated in Table 1.

Table 1: Terminologies for contamination classes on integrated indices

S. No.	Values	Contamination classes
1	1	Unpolluted
2	2	Low polluted
3	3	Moderately polluted
4	4	Strongly polluted
5	5	Extremely polluted

When the PI_{Avg} and $PI_{vectorM}$ are used with the $\Sigma wi=1$ condition, terminologies can also be used like single indices. ΣTr or ΠTr would be used for the integrated indices based on the single index.

Index Comparison

Contamination indices measure the contamination or enrichment levels and ecological risk indices; evaluate the potential for observing adverse biological effects. For index performance evaluation the indices were scored on the basis of qualitative expert knowledge and judgment (the project research team), using the following criteria:

- Comparability: the existence of a target level or threshold against which to compare it so that users are able to assess the significance of the values associated with it.
- Representativity: ability to provide a spatially representative picture of estuarine environmental states and impacts.
- Credibility: a good theoretical basis in technical and scientific terms; applicability to estuaries.
- Simplicity: ease of calculation and interpretation.
- Sensitivity and robustness: responsiveness to change in the environment. vi. Acceptable levels of uncertainty.

Each index was scored from 1 (lowest performance) to 3 (highest performance) for every criterion presented above, and a total performance score was summarized for all the indexes used (table 2).

In each management unit the indices were calculated using the median values of chemical concentration in all the locations belonging to each management area. This mode was also used where the index was nominal. These measures of the central tendency were used instead of an arithmetic mean as the objective of the analysis is to show the main trend in the index values for each management area. Moreover, the arithmetic mean should only be used for normal distributions and should not be used in the present of outliers (Wheater and Cook, 2002).

For MPI and PLI a geometric increment was employed which was divided into four classes. MPI used a classification from clean to highly contaminated (as it is only a contamination index); for PLI a classification from unimpacted to highly polluted was given, according to the index author's classification.

In an overall comparison of the contamination and ecological risk indices the MPI and PLI indices have the highest performance scores, according to the indicator criteria MPI due to its simplicity and PLI due to its simplicity, representative, comparability, sensitivity and robustness. It has the lowest performance score since it needs reference site values. It may give imprecise

Table 2 Score of the metal assessment indices, based on several criteria

	Contamination indices		Ecological risk indice (PLI)
	MPI	I	
Simplicity	3	3	3
Representative	1	2	3
Credibility	2	3	2
Comparability	1	1	3
Sensitivity and robustness	1	3	3
Acceptable levels of uncertainty	2	2	2
Total	16	14	16

Table 3: Pollution index calculated at three depths using average, vector modulus and Nemerow pollution indices

S. No.	Indices	Depth cm		
		0	5	10
1	Average of pollution index	1.08	1.28	1.21
2	Vector modulus of pollution index	1.08	1.29	1.24
3	Nemerow pollution index	1.16	1.37	1.04

Table 4: Pollution index for soil near sugar mill at three depths (0 cm, 5 cm and 10 cm)

Metals	Depth	W_i	C_i	W_iC_i
Fe	0 cm	0.05	20.10	1.01
	5 cm	0.09	15.44	1.39
	10 cm	0.10	16.86	1.69
Mn	0 cm	0.16	7.66	1.23
	5 cm	0.20	7.24	1.45
	10 cm	0.17	6.60	1.12
Zn	0 cm	0.18	5.87	1.06
	5 cm	0.26	4.10	1.07
	10 cm	0.11	9.21	1.01
Cu	0 cm	0.14	7.31	1.02
	5 cm	0.21	5.80	1.22
	10 cm	0.10	10.11	1.01

values because of the undue influence of one of the measurements used in the final composite values (DelValls *et al.*, 1998). It has no threshold for maximum pollution and does not allow comparison between ecosystems.

RESULTS AND DISCUSSION

Average, vector modulus of pollution index and Nemerow pollution index calculated at three depths were tabulated in table 3.

The results of average and vector modulus pollution indices show that the area was unpolluted where as Nemerow pollution index categorizes the region as slightly polluted domain. The values of Nemerow were slightly larger than those of average and vector modulus and the latter two were almost consistent (Gong Qingjie *et al.*, 2008).

Table 5: Pollution load index for soil near sugar mill at three depths (0 cm, 5 cm and 10 cm)

Metals	Depth	C	T	B	PLI
Fe	0 cm	14.78	2.96	7.54	0.26
	5 cm	10.71	6.00	2.88	0.03
	10 cm	10.34	5.26	2.84	0.01
Mn	0 cm	6.42	5.75	12.82	1.24
	5 cm	5.24	4.25	3.84	0.00
	10 cm	5.84	5.15	2.90	0.49
Zn	0 cm	5.29	3.76	3.14	0.00
	5 cm	3.70	3.12	1.80	0.36
	10 cm	8.19	5.86	0.76	0.35
Cu	0 cm	6.74	5.62	1.74	0.51
	5 cm	4.83	4.22	2.12	0.51
	10 cm	8.22	1.86	1.44	0.00

Table 6: Index for chemistry for soil near sugar mill at three depths (0 cm, 5 cm and 10 cm)

Metals	Depth	V_i	$(V_i)_o$	$RTR_i = V_i/V_{io}$
Fe	0 cm	14.78	7.54	1.96
	5 cm	10.71	2.88	3.72
	10 cm	10.34	2.84	3.64
Mn	0 cm	6.42	12.82	0.50
	5 cm	5.24	3.84	1.36
	10 cm	5.84	2.90	2.01
Zn	0 cm	5.29	3.14	1.68
	5 cm	3.70	1.80	2.06
	10 cm	8.19	0.76	10.78
Cu	0 cm	6.74	1.74	3.87
	5 cm	4.83	2.12	2.28
	10 cm	8.22	1.44	5.71

The pollution index calculated was tabulated in table 4 which shows that the surface soil was strongly polluted (PI_{0cm} = 30.78) and at depths 5 cm and 10 cm were moderately polluted (PI_{5cm} = 10.26 and PI_{10cm} = 9.66 respectively).

Pollution load index which indicates the ecological risk index predicted that the surface soil was moderately polluted compared to depths of 5 cm and 10 cm which were unpolluted and the result was in accordance with the findings of Sayadi *et al.*, (2009). The PLI values were calculated to be 4.02, 1.80 and 1.70 respectively (table 5).

Indices for chemistry (I) at three depths were 16.02, 18.84 and 44.28 respectively. According to the classes categorized depths 0 cm and 5 cm were strongly polluted and depth of 10 cm was extremely polluted where as results of metal pollution index predicts that all

Table 7: Metal enrichment index for soil near sugar mill at three depths (0 cm, 5 cm and 10 cm)

Metals	Depth	Ci	Co	SEF
Fe	0 cm	14.78	7.54	0.96
	5 cm	10.71	2.88	2.72
	10 cm	10.34	2.84	2.64
Mn	0 cm	6.42	12.82	-0.49
	5 cm	5.24	3.84	0.36
	10 cm	5.84	2.90	1.01
Zn	0 cm	5.29	3.14	0.68
	5 cm	3.70	1.80	1.06
	10 cm	8.19	0.76	9.78
Cu	0 cm	6.74	1.74	2.87
	5 cm	4.83	2.12	1.28
	10 cm	8.22	1.44	4.71

Table 8: Potential ecological risk index for soil near sugar mill at three depths (0 cm, 5 cm and 10 cm)

Metals	Depth	C_i	C_{SQV}	ERF
Fe	0 cm	14.78	20.10	-0.26
	5 cm	10.71	15.44	-0.31
	10 cm	10.34	16.86	-0.39
Mn	0 cm	6.42	7.66	-0.16
	5 cm	5.24	7.24	-0.28
	10 cm	5.84	6.60	-0.12
Zn	0 cm	5.29	5.87	-0.09
	5 cm	3.70	4.10	-0.09
	10 cm	8.19	9.21	-0.11
Cu	0 cm	6.74	7.31	-0.08
	5 cm	4.83	5.80	-0.17
	10 cm	8.22	10.11	-0.19

the three depths were extremely polluted with $MPI_{0,\ 5\ and\ 10\ cm}$ = 7.63, 5.63 and 7.98 respectively (table 6). These findings were in accordance with the results of Bakkialakshmi and Vinodhini (2008) as they reported high contamination of the heavy metals in soils near sugar mill.

The metal enrichment index categorized different depths as moderately and extremely polluted with $SEF_{0,\ 5\ and\ 10\ cm}$ = 10.00, 10.84 and 36.28 respectively (table 7) where as potential ecological risk index categorizes the three depths as unpolluted (Bhupander Kumar *et al*., 2011) (table 8).

Soil Quality Near Sugar Mill

Average and vector modulus predict that the region near sugar mill was unpolluted where as Nemerow pollution index classify the area as slightly polluted domain. Most of the integrated indices predicted the place as strongly polluted. These results indicate that the soil quality varied from strongly polluted to unpolluted as the depth increases. This lack of consistent correlation of the anthropogenic metals in the slightly deeper soil layers may indicate that there is minimal bioaccessibility and/or bioavailability of these metals within the region, an observation not necessarily consistent with the conjecture of Nriagu *et al*., (1998) about long term release of metals to regional waters as a result of soil weathering.

Geochemical carriers

Pearson correlation matrix, standard deviation and variance for analyzed soil parameters were calculated to determine the interrelation between the parameters (table 9 and 10). Examination of the matrix also provides clues about the carrier substances and the chemical association of trace elements in the studied areas (Forstner, 1981; Jaquet *et al*., 1982).

Standard deviation measures the absolute dispersion of a distribution. A small standard deviation means a high degree of uniformity in the observations as well as homogeneity of the series (P.N.Arora *et al*, 2007). Variance has a great practical significance and is the best measure of comparing the variability of the series. Except iron all the metals show

Table 9: Statistical data of heavy metals near sugar mill

Metals	Depth	Mean	Minimum	Maximum	Standard deviation	Variance
Fe	A (0 cm)	14.78	2.96	20.1	7.41	54.93
	B (5 cm)	10.71	6.00	15.44	3.45	11.89
	C (10 cm)	10.34	5.26	16.86	4.98	24.79
Mn	A (0 cm)	6.42	5.75	7.66	0.79	0.64
	B (5 cm)	5.24	4.25	7.24	1.00	1.01
	C (10 cm)	5.84	5.15	6.60	1.41	1.98
Zn	A (0 cm)	5.29	3.76	5.87	0.86	0.75
	B (5 cm)	3.70	3.12	4.10	3.59	12.91
	C (10 cm)	8.19	5.86	9.21	1.32	1.75
Cu	A (0 cm)	6.74	5.62	7.31	0.65	0.42
	B (5 cm)	4.83	4.55	5.80	2.03	4.13
	C (10 cm)	8.22	1.86	10.11	1.28	1.64

Table 10: Correlation coefficient matrix between heavy metal concentration in soil near sugar mill

	Fe	Mn	Zn	Cu
Fe	1	-0.430	0.189	0.282
Mn		1	0.018	0.020
Zn			1	0.808
Cu				1

homogeneity in values with depth. Iron shows high variation with depth.

Iron showed negative correlation (-0.430) with Manganese which indicate that increase in the concentration of iron will lead to decrease in the concentration of manganese. Zinc exhibited significant positive correlation with Copper which indicate that the presence of Zinc in the soil will lead to the accumulation of Copper. This result is at variance with the report of Aikpokpodion *et al.*, (2010) where existed positive correlation between large number of heavy metals.

CONCLUSION

Different metal assessment indices were used and discussed. Some indices gave equivalent information but others gave complementary information (e.g. contamination or background enrichment indices and ecological risk indices) that can be developed for different purposes. There should be better methods of standardization for indices to allow better comparability between them (as several assess the same information).

According to the evaluation of the index criteria performance, PLI had the highest score particularly in the group of ecological risk indices. This index can be complemented with the MPI contamination index. MPI does not evaluate the potential for adverse effects and the results from one ecosystem are more difficult to compare with others, but it allows more site-specific and accurate information on contamination levels. The results of the indices per management unit are in accordance with the surface areas of each metal. If the aim of contamination evaluation is to assess the overall contamination of a study area, the indices are highly appropriate.

Heavy metal assessment indices are not to be used as the only evidence of soil quality. In future developments, organic compounds will be integrated into the contamination evaluation, which can be correlated with data on the different sources and spatial distribution of pollution. Furthermore, the integration of contamination assessment with biota and toxicity evaluation will be carried out in each management unit to allow a weight of evidence for soil quality assessment near sugar mill.

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