Assessment of Heavy Metal Pollution in Abandoned Coal Stockpile and Lime Kiln of Meghalaya using Pollution Load Index (PLI) and Geoaccumulation Index (I-Geo)

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Abstract: Soil pollution problems from potential toxic heavy metals have been a serious concern nowadays due to anthropogenic activity like mining and industrial activity. Thus, the present study was aimed to assess the pollution of soil by heavy metals in abandoned coal stockpile and abandoned lime kiln by using pollution indices calculation. The results revealed that the soils are low in organic contents (N, P, K and C) in both the study sites. The heavy metal concentration in the soil of both the study sites are in the order Fe>Mg>Zn>Mn>Se>Na> As>Ca>Pb>Cu>Ni=Cr>Co>Li in abandoned coal stockpile and Ca>Fe>Mg>Se>As>Na>Mn>Zn>Pb>Cu>>Cr>Ni>Co in abandoned lime kiln. Based on the CF values indicate that the soils were contaminated with Se, Fe in abandoned coal stockpile and Se, Fe and Ca in abandoned lime kiln. The degree of contamination shows a high contamination in both the study sites. The modified degree of contamination suggest that is moderately contaminated $(mC_d=2.23 \text{ and } 3.14)$ in both the study sites. Pollution load index were found to be generally low (<1) and geoaccumulation index revealed that the soil are strongly contaminated by Se, moderately to strongly contaminated by Ca in abandoned lime kiln, moderately contaminated by Fe. This study gave us a baseline understanding on soil contamination by heavy metals in abandoned sites by using these pollution indices.

Keywords: Contamination Factor, Degree of Contamination, Pollution Load Index, Geoaccumulation Index, heavy metals

1. Introduction

In the recent past, many countries having mining industries are facing soil pollution problems from potential toxic heavy metals especially from abandoned mine lands. Heavy metals occur naturally as a constituent of the Earth crust. At trace levels, some of these heavy metals are biologically essential for all the living organisms especially in human health (Juvanovic et al., 1995). Metallic mining have resulted to elevated heavy metals accumulation in the soil and cause a potential risk to the soil health environment and are detrimental to population (Abrahams, 2002). Mining activities generally results in the deposition of very large significant volumes of wastes on soil and inturn to plants and animals exposed to the elements contained in the residue. The direct effect of mining is the overall loss of production due to the loss of forest, cultivated or grazing land and the indirect effects include air and water pollution and siltation of water body which will eventually lead to

loss of biodiversity, amenity and economic wealth (Yang et al., 2002; Wong, 2003).

Meghalaya is endowed with great minerals wealth and coal and limestone are the two important minerals where they are mining have been carried out. However, mining of coal have been done in a very unscientific way and absence of post mining treatment and management of mined areas have lead

to large scale land cover/land use changes degradation in the mining areas. The dumping of coal near the unmine site generates huge quantity of mine spoils or overburden in the form of gravels, rocks, sand, soil etc which destroys the surrounding vegetation and leads to severe soil and water pollution. The small-scale limekiln sector is an important contributor to the economic development and for rural employment in the state. Most of the limekilns are situated in the proximity of limestone deposits. Most of the kilns employ outdated technologies with very low energy efficiency, low yield, and poor environmental performance. Limekilns activities also caused severe degradation of vegetation and vegetation floor as well as pollution of the streams due to the runoff from these kilns during heavy rains. Therefore, the objective of the present study was to assess the concentration of heavy metals in the soils of these two abandoned sites by using pollution load index and geoaccumulation index.

2. Materials and methods

2.1 Study Site

In this study, an abandoned coal stockpile and lime kiln in Cherrapunjee (Fig 1) was selected for study. The soil samples were collected from the study sites during the month of May, 2010

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Figure 1: Map showing study sites

2.2 Sampling technique and Preparation

Surface soil was randomly sampled using hand driven steel corers up to the depth of 10 cm and collected in labeled polythene bags and then they are transferred to the laboratory for pre-treatment and analysis. In the laboratory, bulk soil samples were spread on travs and were air dried for 15 days at room temperature. The samples were then grounded using mortar and pestle and sieved through a 2 mm mesh and homogenized and stored in polyethylene bags before used. For heavy metal analysis, microwave digestion was performed in Perkin Elmer Microwave digester 3000. 0.5 g of the soil sample were weighed into a 120-mL microwave digestion vessel, then 8 mL of freshly prepared aqua regia solution $(1/3 \text{ HNO}_3\text{-HCl}, v/v)$ was added and the samples were digested for 5 min, then filtered through Whatman filter paper no. 42, transferred to 100 mL volumetric flasks, and diluted with deionized, distilled water. The metals (Cu, Fe, Mn, Co, Pb, Cr, Ni, Se, Zn, Ca, Mg, Na, Li, and As) in the soil extracts were analyzed by Atomic Absorption Spectrometry (AAS 3110, Perkin Elmer). The pH and EC of the soil samples were measured using pH and EC meters (Decibel, India), Available phosphorus from soil was determined following molybdenum-blue method (Allen et al. 1974) and soil organic carbon was estimated using colorimetric method (modified Walkey and Black method) and nitrogen by the Kjeldahl method.

2.3 Pollution Indices

Geo-accumulation index (I-geo) and Pollution Load Index (PLI) were employed to assess the pollution of metals in these two abandoned sites.

1. Geo-accumulation index (I-geo)

To assess the level of contamination in soils Geoaccumulation Index was used to calculate the changes in the original pre-industrial metal concentrations and the current levels of metal concentrations in the soils. Geoaccumulation Index approach as proposed by Muller, (1979) was used as an assessment on the effects of mining activities. The formula used for the calculation of Igeo is expressed as follows:

$$I_{geo} = \log_2\left(\frac{Cn}{1.5xBn}\right)(1)$$

Here, Cn is the measured concentration in the sediment for metal 'n', Bn is the background value for the metal 'n' (Turekian and Wedepohl, 1961) and the factor 1.5 is used to minimized the effect of possible variations in the background data, Bn which may be attributed to lithological variations in soils.

2. Concentration Factor

The contamination factor can be calculated from the following relation:

$$CF = \frac{Metal \ concentration \ in \ t \square e \ sample}{Background \ value \ of \ t \square e \ metal} (2)$$

3. Modified Degree of Contamination mCd

The degree of contamination in soil by metals was calculated based on the method presented by Likuku et al., (2013) as shown below.

$$mC_{d} = \sum_{i=1}^{n} CFi(3)$$

Where N is the number of elements analysed and CF is the contamination factor calculated as shown in (2)

$$C_{d} = \sum_{i=1}^{N} CFi....(4)$$

The aim of calculating contamination factor is to provide a measure of the degree of overall contamination of surface layers in a sampled site. Furthermore, all the metals analysed in order to determine the correct C_d that would satisfy classification technologies specified by Hakanson, (1980).

4. Pollution Load Index (PLI)

The pollution load index, proposed by Thomlinson et al., (1980) was calculates using (5)

$$PLI = \sqrt[N]{(CF1 \ X \ CF2 \ X \ CF3 \ X \ \dots \ X \ CFN \ (5))}$$

Where N is the number of metals studied and CF is the contamination factor calculated as described in (2). PLI provides a simple, comparative means for assessing a site- a value of zero indicates perfection, a value of one indicates only baseline levels of pollutants present and values above one would indicate progressive deterioration of the site (Tomlinson et al., 1980).

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Statistical analysis

Correlation of different elements in soil samples were calculated by Pearson correlation matrix and ANOVA was performed for all the data to confirm their validity using SPSS 17. Data were presented as mean±standard error of three replicates.

3. Results and Discussion

The soil pH values in the abandoned lime kiln is basic in nature at 9.86 this maybe due to presence of large amounts of exchangeable Ca and Mg (Boateng et al., 2012) as compared to the abandoned coal stockpile where the is acidic in nature at 5.23 (Table 1). The soils EC values of both the abandoned sites are less than 1 s/m. Organic carbon and available nitrogen contents are very low, similarly available phosphorus is less than 1 mg/kg in both the study and available K levels were low (<4 mg/kg soil) in both the study. The probable reasons of less organic contents in both

the study sites may be due to the dumping of the mining materials on the surface soils which leads to the loss of these organic contents.

Table 1: pH,	conductivity	and	organic	contents	measured	in
	the	stud	v sites			

Sites	Parameters										
	pН	С									
		(S/m)	(mg/kg)	(mg/kg)	(mg/kg)	(mg/kg)					
Lime Kiln	9.86±	$0.48\pm$	113.23±	0.024±	1.4±	0.34±					
	0.003	0.001	6.5	0.1	0.1	0.01					
Coal stockpile	5.23±	0.49±	50.93±	0.012±	3.45±	0.68±					
-	0.03	0.1	3.2	0.1	0.1	0.1					

3.1 Concentration of the Studied Metals

Concentration of Cu, Fe, Mn, Co, Pb, Cr, Ni, Se, Zn, Ca, Mg, Na, Li, and As in the abandoned coal stockpile and lime kiln sites are presented in Table 1.

Table 2: Statistical data of the 14 metal	ls (mg/kg) measured in this study
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Metals		Lime Kiln		Coal stockpile						
	(min-max)	Mean	p-value	(min-max)	Mean	p-value				
Cu	(0.22-0.26)	0.24±0.01	0.003	(0.2-0.21)	0.209±0.1	0.000				
Fe	(23.57-23.77)	23.67±0.05	0.000	(25.14-25.18)	25.16±0.1	0.000				
Mn	(0.79-0.79)	0.79±0.01	0.000	(1.33-1.334)	1.33±0.1	0.000				
Co	(0.09-0.13)	0.11±0.01	0.011	(0.10-0.12)	0.11±0.1	0.003				
Pb	(0.31-0.33)	0.32±0.1	0.000	(0.24-0.35)	0.29±0.1	0.012				
Cr	(0.15-0.16)	0.16±0.1	0.000	(0.14-0.16)	0.15±0.1	0.001				
Ni	(0.1-0.15)	0.12±0.1	0.013	(0.06-0.2)	0.15±0.1	0.072				
Se	(1.2-1.79)	1.49±0.17	0.013	(0.18-2.29)	1.22±0.6	0.182				
Zn	(0.38-0.41)	0.4±0.1	0.000	(1.49-1.75)	1.61±0.1	0.002				
Ca	(30.62-34.29)	32.54±1.06	0.001	(0.37-0.38)	0.37±0.1	0.000				
Mg	(2.03-2.07)	2.05±0.01	0.000	(1.98-2.18)	2.08±0.1	0.001				
Na	(1.04-1.07)	1.05±0.01	0.000	(1.17-1.22)	1.2±0.1	0.000				
Li	ND	ND	-	(0.04-0.05)	0.05±0.1	0.004				
As	(1.28-1.29)	1.28±0.1	0.000	(0.90-0.93)	0.91±0.1	0.000				

The results also shows that the heavy metal concentration in the soils of both the study sites are in the order Ca>Fe>Mg>Se>As>Na>Mn>Zn>Pb>Cu>>Cr>Ni>Co in abandoned lime kiln and Fe>Mg>Zn>Mn>Se>Na> As>Ca>Pb>Cu>Ni=Cr>Co>Li in abandoned coal stockpile. The mean concentrations of Fe, Mn, Zn and Mg in abandoned coal stockpile exceeded (p<0.05) than those in abandoned lime kiln. Elevate differences in mean concentrations of these 4 metals in the abandoned coal stockpile maybe due to the deposition of toxic substances associated with dumping of coal in the surface soils. On the other hand, Ca, Se and As concentrations in the abandoned lime kiln were higher as compared to abandoned coal stockpile. The high concentration of Ca, Se and As in the soils from the abandoned lime kiln are statistically higher (p<0.01) than those found in soil samples from the abandoned coal stockpile. The high mean concentration of Ca in abandoned lime kiln maybe due to a direct influence of industrial activity as an anthropogenic input in calcium concentration in the soils of the surrounding area. The insignificant mean differences in Fe concentration in both the study sites maybe due to its natural abundance.

3.2 Contamination Factors, Degree of Contamination, Modified Degree of Contamination and Pollution Load Index

Soils samples in both the sites i.e., abandoned coal stockpile and abandoned lime kiln were also assessed for contamination factors, degree of contamination, modified degree of contamination and pollution load index are given in Table 3 and 4.

Table 3: Contamination factors, degree of contamination, modified degree of contamination and pollution load index of the studied metals in abandoned coal stockpiled

	studied metals in abandoned coal stockplied													
	Cu	Fe	Mn	Co	Pb	Cr	Ni	Se	Zn	Ca	Mg	Na	Li	As
CF	0.003	4.46	0.001	0.004	0.023	0.001	0.001	24.7	0.023	0.09	0.89	0.5	0.001	0.5
C_d	31.23													
mC_d	2.23													
PLI	0.04													

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 Table 4: Contamination factors, degree of contamination, modified degree of contamination and pollution load index of the studied metals in abandoned Limestone kiln

	Cu	Fe	Mn	Co	Pb	Cr	Ni	Se	Zn	Ca	Mg	Na	Li	As
CF	0.004	4.20	0.0008	0.004	0.025	0.001	0.001	29.9	0.005	7.82	0.87	0.44	ND	0.71
C_d	44.01													
mC_d	3.14													
PLI	0.074													

The assessment of soil contamination was carried out using the contamination factor and the degree of contamination, based on four classification categories recognized by Hakanson, (1980) where the modified degree of contamination, mC_d classification and description (Abrahim and Parker, 2008) are given in Table 5 and 6 respectively.

 Table 5: Contamination factors and degree of contamination categories and terminologies

CF classes	CF and C _d terminologies	C _d classes
CF<1	Low CF indicating low	C _d <8
	concentration/low C _d	
1≤CF<3	Moderate CF/ C _d	$8 \le C_d < 16$
3≤CF<6	Considerable CF/ C _d	$16 \le C_d < 32$
CF>6	Very high CF/ C _d	$C_d \ge 32$

 Table 6: Modified degree of contamination classification and description

mC _d classes	Modified degree of contamination level
$mC_d < 1.5$	Nil to very low degree of contamination
$1 \le mC_d < 2$	Low degree of contamination
$2 \le mC_d < 4$	Moderate degree of contamination
$4 \le mC_d < 8$	High degree of contamination
$8 \le mC_d < 16$	Very high degree of contamination
$16 \le mC_d < 32$	Extremely high degree of contamination
$mC_d \ge 32$	Ultra high degree of contamination

The overall contamination of soils in abandoned coal stockpile, based on the CF values indicate that the soils were contaminated with Se, considerably contaminated by Fe and shows signs of low contamination of other metals. In abandoned lime kiln the CF values indicates that the soil was contaminated with Se and Ca, considerably contaminated by Fe and low contamination of other metals. The degree of contamination shows a high contamination in both the study sites. The modified degree of contamination suggest that is

moderately contaminated (mC_d= 2.23 and 3.14) in both the study sites.

Pollution load index (PLI) was used to determine the pollution severity and its variation along the study sites and also it can be used as a tool to compare the pollution status of different places (Rabee et al., 2011). Results of the present study showed that Pollution Load Index (Table 3 and 4) were found to be generally low (<1) in both the study sites. These confirmed that the abandoned coal stockpile and lime kiln is facing probable environmental pollution especially with dangerous heavy metals (Se and As).

3.3 Geoaccumulation Index

The index of geoaccumulation was assessed based on the seven descriptive classes for increasing I_{geo} value proposed by Muller, (1969) depicted in Table 6.

Table 7: Geoaccumulation Index degree of contamination
classification and description

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Value	Class	Description										
$I_{geo} > 5$	6	Extremely contaminated										
$4 < I_{geo} \le 5$	5	Strongly to extremely contaminated										
$3 < I_{geo} \le 4$	4	Strongly contaminated										
$2 < I_{geo} \leq 3$	3	Moderately to strongly contaminated										
$1 < I_{geo} \le 2$	2	Moderately contaminated										
$0 < I_{geo} \le 1$	1	Uncontaminated to moderately										
5		contaminated										
$I_{aeo} = 0$	0	Uncontaminated										

The geoaccumulation index values (Table 7) for the abandoned coal stockpile and abandoned lime kiln revealed that the soils are strongly contaminated by Se, moderately to strongly contaminated by Ca in abandoned lime kiln, moderately contaminated by Fe and uncontaminated by other metals in both the abandoned study sites.

	rable 0. igeo values													
	Cu	Fe	Mn	Co	Pb	Cr	Ni	Se	Zn	Ca	Mg	Na	Li	As
Lime Kiln	-8.41	1.48	-10.8	-8.34	-5.87	-9.85	-9.79	4.31	-8.03	2.38	-0.76	-1.74	0	-1.07
Coal stockpile	-8.62	1.57	-10.06	-8.31	-5.99	-9.89	-9.7	4.04	-6.01	-4.05	-0.74	-1.55	-1.55	-1.55

The negative values in Cu, Mn, Co, Pb, Cr, Ni, Zn, Mg, Na As in both the abandoned coal stockpile and lime kiln and Ca and Li in abandoned coal stockpile indexes of geoaccumulation shown in Fig 2 and 3 may be the results of low levels of contamination in the soil samples.

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Fig 2: Contamination Factor and geoaccumulation factor in abandoned coal stockpile



Fig 3: Contamination Factor and geoaccumulation factor in abandoned lime kiln

3.4 Correlation Analysis

Pearson's correlation coefficients for Cu, Fe, Mn, Co, Pb, Cr, Ni, Se, Zn, Ca, Mg, Na, Li, and As in the abandoned coal stockpile and lime kiln sites are depicted in Table 9 and 10. The matrix summarizes the strength of the linear relationships between each pair of variables. All the metal pairs in the soil samples exhibit positive relations (except for the Cu-Mn, Fe-Mn, Co-Pb, Co-Cr, Co-Zn, Cr-Ca, Se-ca, Se-Na, Mg-Na, Mg-As pairs) in abandoned coal stockpile and Fe-Co, Fe-Na, Co-Mn, Cr-Ca, Se-Zn, Se-Ca, Mg-Na pairs in abandoned lime kiln, but some of them were significant at the 95%, 99% and 100% confidence levels. The Cu-Fe, Cu-Ca, Cu-Na, Cu-As, Fe-Se, Fe-Ca, Fe-Na, Fe-As, Pb-Cr, Pb-Zn, Cr-Zn, Ca-As and Na-As pairs in abandoned coal stockpile sites and Fe-Mn, Co-Na, Pb-Ni, Cr-Se, Zn-Na pairs in abandoned lime kiln sites are significantly positively correlated which may suggest a common pollution source or a similar geochemical behavior for these metals and Pb-Zn pair maybe due to vehicular emissions as in abandoned coal stockpile trucks are used to transport the mining coal materials. Negative and inverse correlations between metals indicate that these metals are derived from different sources and that this metal is not associated with other metals.

	Cu	Fe	Mn	Со	Pb	Cr	Ni	Se	Zn	Ca	Mg	Na	Li	As
Cu	1													
Fe	1.000**	1												
Mn	.000	.000	1											
Co	.982	.982	189	1										
Pb	988	988	.156	999*	1									
Cr	992	992	.127	998 *	1.000*	1								
Ni	.928	.928	373	.982	975	968	1							
Se	-1.000*	-1.000*	.019	985	.991	.994	935	1						
Zn	980	980	.200	-1.000**	.999*	.997*	984	.983	1					
Ca	.998*	.998*	064	.992	996	998 *	.950	999 *	991	1				
Mn	998 *	998 *	063	968	.976	.982	902	.997	.965	992	1			
Na	.999*	.999*	.048	.972	979	985	.909	998 *	969	.994	-1.000**	1		
Li	990	990	143	945	.955	.964	865	.987	.941	979	.997	995	1	
As	1.000**	1.000**	.000	.982	988	992	.928	-1.000*	980	.998*	998 *	.999*	990	1

Table 9: Pearson	correlation	matrix	of abandoned	coal stockpile
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*. Correlation is significant at the 0.05 level (2-tailed).

**. Correlation is significant at the 0.01 level (2-tailed).

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	Table 10. 1 curson conclution matrix of abababiled mile kin												
	Cu	Fe	Mn	Со	Pb	Cr	Ni	Se	Zn	Ca	Mg	Na	As
Cu	1												
Fe	.979	1											
Mn	.318	.120	1										
Со	977	-1.000**	109	1									
Pb	852	940	.225	.944	1								
Cr	.926	.983	064	985	987	1							
Ni	871	952	.189	.955	.999*	992	1						
Se	.945	.992	010	993	977	.999*	984	1					
Zn	954	995	019	.996	.970	997	.978	-1.000*	1				
Ca	917	979	.086	.981	.990	-1.000*	.995	997*	.994	1			
Mn	.978	1.000**	.115	-1.000**	942	.984	954	.992	995	980	1		
Na	973	-1.000*	091	1.000^{*}	.950	988	.961	995	.997*	.984	-1.000*	1	
As	792	899	.327	.903	.994	964	.990	948	.939	.970	901	.911	1

Table 10: Pearson correlation matrix of ababdoned lime kiln

*. Correlation is significant at the 0.05 level (2-tailed).

**. Correlation is significant at the 0.01 level (2-tailed).

4. Conclusion

Our investigations revealed that soil samples in the two abandoned study sites that organic contents (C, N, P and K) in the soil are very low. Assessment of soils pollution by heavy metals in the abandoned coal stockpile and abandoned lime kiln was evaluated using contamination factor, modified degree of contamination factor, pollution load index and geoaccumulation index. The contamination of soils at was study sites revealed that the soils were contaminated with Se, in the abandoned coal stockpile and Ca and Se in the abandoned lime kiln. The degree of contamination shows a high contamination of metals in the of both the study sites. The modified degree of contamination which determines an average value of individual CF values of the studied metals revealed a moderately contaminated (mC_d= 2.23 and 3.14) by the studied metals in the study sites. The pollution load index suggest that the contamination of the soils by these metals is less (PLI<0.5) which is generally very low pollution, The Igeo calculations revealed that these two abundant sites are strongly contaminated by Se followed by Ca and Fe. This study gave us a baseline understanding on soil contamination by heavy metals in abandoned sites by using these pollution indices, however detailed study is needed to monitor these sites in order to prevent further soil contamination by heavy metals and possible remediation of these abandoned sites.

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