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Assessment of Heavy Metal and Pollution Indices in Rubber Plantation Soil: A Case Study in Jeli, Kelantan, Malaysia

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ABSTRACT: The present study investigated the concentrations of 12 selected metals in soils collected from a rubber plantation area located at Jeli, Kelantan, The collection of soils was conducted during wet and dry seasons. The total concentrations of the selected metals were treated by using X-ray fluorescence spectrometer (XRF). A comparison of the soils between the two seasons showed that Si was highly accumulated in soils for dry (631000-761000 mg/kg) and wet (726000-796000 mg/kg) seasons, while Pb (140-153 mg/kg) and Zn(135-170 mg/kg) were detected as the lowest accumulation in soils for dry and wet seasons, respectively. However, Al was detected second highest after Si metal with mean concentration ranged from 152000-230000 mg/kg (dry season) and 130000-167000 mg/kg (wet season), followed by K (10600-22000 mg/kg in dry season and 5930-19400 mg/kg for wet season), Ti (9080-14900 mg/kg in dry season and 7980-11300 for wet season). Zr (4050-5140 mg/kg in dry season and 5990-7240 mg/kg in wet season), Cl (120-881 mg/kg in dry season and 677-2110 mg/kg in wet season), Mn (351-602 mg/kg in dry season and 33-548 mg/kg in wet season), Cr (282-395 mg/kg in dry season and 232-308 mg/kg in wet season), and Sn (150-172 mg/kg in dry season and 176-199 mg/kg in wet season). The obtained results reflect the higher levels of metals accumulated in soils collected from dry season, when compared to wet season. In comparison, the Enrichment Factor (EF), the Geo-accumulation Index (I-geo), and the Pollution Load Index (PLI) suggested that these metals have the potential to cause environmental risks. Nevertheless, prevention measures can be executed to reduce the impacts of heavy metals.

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1. Introduction

Metal contamination in soil has been known as a critical environmental issue and has become a worldwide concern as trace metals are nonbiodegradable and persistent.¹ Moreover, metals can produce toxicant effects when present at high levels in soil as a result of agricultural activity through application of fertilisers and pesticides². Apparently, industry, municipal waste, vehicular emission, and organic manure $^{3-6}$ are collectively accounted for higher concentration of toxicants in soil. According to Desaules ⁷ anthropogenic trace metals mostly deposited on the soil surface as agricultural soils are the most potential contaminants introduced into the soil and mainly derive from human activities, as well as deposition by the application of agrochemicals⁸. These potential toxic elements could accumulate slowly in the soil profile over long periods of time. The agriculture sector is the third important sector that contributes to

the economic growth after the manufacturing and service sectors in Malaysia ⁹. Various studies of metals pollution in agricultural lands have been conducted, for example, a study of heavy metals in agricultural soils from Cameron Highland, Pahang, and Cheras, Kuala Lumpur ¹⁰, Kedah, and Penang ¹¹. Toxicant metals have the potential to accumulate, which may affect the end users.

In this study, Jeli District was selected as the study site as this area is potentially contaminated by metals through rubber planting activity. Hence, it is appropriate to conduct a study on metal pollution in soil at Jeli since there is lack of study that had looked into the heavy metals in soil within this area. Assessment of soil pollution can be accomplished through various methods, in which the common ones are Enrichment Factor (EF), Geo-accumulation Index (I-Geo), and Pollution Load Index (PLI) ¹²⁻¹⁴. The objectives of this study are to determine the

concentrations of selected metals in soil samples and to assess the metal pollution in soil through pollution indices.

2.0 Methods and Materials

2.1 Study Area

Two rubber plantations in Jeli were selected as sampling sites; Kampung Gemang and Kampung Kulim, which were geo-located using geographical positioning system (GPS), as illustrated in Table 1 and Figure 1. Jeli District was selected as the study area because this area is potentially contaminated due to its agricultural activity, which is rubber plantation. The soil samples were collected at two points at each of the selected rubber plantation, thus making it four points.

2.2 Sample Collection and Preparation

2.2.1 Soil sampling

The soil samples were collected at two stations using auger with a depth of 10 cm. The soil samples were placed in polyethylene plastic bags and labelled. Next, the samples were preserved in a covered cooler with ice at approximately 4 °C, wrapped with aluminium foil, transported back to the laboratory, and stored in the freezer at -20°C for further analysis. The soil samples were dried in an air-circulating oven at 80 °C for 3 days. Soon after drying, the samples were crushed into powder form using a mortar and pestle and sieved through 2mm stainless steel aperture sieve. The samples were sent to the x-ray laboratory and metal concentration was analysed by using an X-ray fluorescence spectrometer (XRF) S2 Ranger model. Portable XRF analysers are able to perform fast because this technique significantly cuts the time required for sample characterisation ¹⁵. The data for metal concentration in the soil samples were recorded in mg/kg unit.

2.2.2 Soil pH

The reading for pH was obtained through in-situ measurement using the twin meter DM.15 model. The pH reading for each point was recorded accordingly.

2.2.3 Moisture Content

The soil samples were placed in an oven at 105 °C and dried until the constant weight was 16. The moisture content was calculated using gravimetric method by applying the following equation:

$$\%W = \frac{A-B}{B} \times 100\%$$

Where:

- %W = Percentage of moisture in the sample,
- A = Weight of wet sample (g), and
- B = Weight of dry sample (g).
- 2.3.4 Total Organic Matter (TOM)

In determining the total organic matter (TOM), the loss-on-ignition method was used. The sediment samples were dried at 70 °C for 24 hours before being combusted at 550 °C for 4 hours in the furnace. According to Bastami et al ¹⁷, TOM can be calculated using the following equation:

Total Organic Matter (TOM, %) = $\left[\frac{B-C}{B}\right] \times 100$ Where,

B = weight of dried sediment before combusting in the furnace

C = weight of dried sediment after combusting in the furnace.

2.3 Statistical Analysis

2.3.1 Assessment of Metal Pollution

The metal pollution indices; EF, I-Geo, and PLI, were utilised in this study.

a) Enrichment Factor (EF)

The value of EF can determine the source of the metal whether from natural or anthropogenic source ¹⁸. The EF is derived from standardisation of a tested element against a reference one ¹⁹. Fe was used as reference element in this study. According to Godwin et al ¹³, EF can be calculated using the following equation:

Where:

[Me/Fe]_{sample} = the metal to Fe ratio in the sample [Me/Fe]_{crust} = the value of metal to Fe ratio in the crust

The value of EF greater than 1.0 indicates anthropogenic origin of the element 13 . The EF was categorised according to Table 2, by adhering to a study conducted by Shaari et al 20 .

b) Geo-accumulation index (I-geo)

The I-geo was used to determine the anthropogenic contamination in the soil. I-Geo was calculated by using the following equation derived from Muller²¹, as shown in equation 2.4:

 $I\text{-}geo = log_2\left[C_n/1.5B_n\right] \qquad)$

Where:

 C_n = the concentration of metals examined in soil samples, and

 B_n = the geochemical background concentration of the metal.

Factor 1.5 refers to possible variations of the background matrix correction factor due to lithological variations ²². I-geo was categorised according to Table 3, as stated by Zahra et al ²³.

c) Pollution Load Index (PLI)

The PLI for each sample was evaluated using the equation derived by Tomlinson et al. $^{\rm 24}$

 $PLI = [CF_1 \times CF_2 \times CF_3 \times CF_n]^{1/n}$

where n = number of metals.

Contamination factor (CF) = Metal concentration in sediment/Background values of the metal ^{13.} (PLI>1) indicates the metal contents in soil are polluted, which can cause harm to the environment and the community, while (PLI<1) indicates unpolluted soil ²⁵.

2. Results & Discussion

2.1 Soil pH, Moisture Content and Total Organic Matter

The summary of soil pH, moisture content, and total organic matter values during dry and wet seasons is given in Table 4. The pH for all stations ranged from 6.2 to 6.7 for dry season and 6.6 to 6.8 for wet season. The soil pH mean recorded in wet and dry seasons were 6.5 and 6.6 respectively, thus, indicating the soil at the studied rubber cultivation site was slightly acidic. According to Verheye ²⁶, the optimal pH for soil in rubber cultivation was between 4.5 and 6.0, however, higher pH values are also tolerated.

The mean value of moisture content during dry season was 32.55 g with the range of 20.20 g to 30.20 g. In wet season, the mean value of moisture content was 36.16 g, ranging between 25.46 g and 51.07 g. These show that the soil samples collected during wet season contained higher moisture contents than that in dry season. The increase in soil moisture content during wet season could be attributed to the amount of rainfall, meanwhile the lower moisture content was resulted from utilisation of water by crops during dry season 27 .

The average of TOM during dry season (0.0005 %) was slightly higher than in the wet season (0.0004 %). It showed a slight decrease in organic matter during the wet season. As stated by FAO ²⁸, the level of soil organic matter usually increases due to increment in annual precipitation as a result of greater biomass production during the rainy season. During rainy season, the amount of organic matter at the top soil also decreases as it is washed away through the runoff ²⁸.

3.2 Metal Concentration in Soil

The total metal concentrations of selected elements during dry and wet seasons are shown in Table 5. Based on the mean values of all metal concentrations in Table 6, the metal concentrations during dry season in descending order is: Si > Al > K > Ti > Zr > Ca > Mn> Cl > Cr > Zn > Sn > Pb. Meanwhile, the descending order of metal concentrations in wet season is: Si > Al > K > Ti > Zr > Ca > Cl > Mn > Cr > Sn > Pb > Zn.

Si metal recorded the highest mean concentration during dry (667500.00 mg/kg) and wet seasons (755750.00 mg/kg) (Figure 1a), ranging from 631000 - 761000 mg/kg in dry season and 726000 -796000 mg/kg in wet season (Table 5). Si is the second most abundant element after oxygen in the earth's crust and it is one of the basic constituents in most soils ²⁹. Meanwhile, Pb and Zn metals had the lowest mean concentration of metals during dry and wet seasons, respectively. The second most abundant metal found in the soil samples was Al. The concentration of Al ranged from 152000 - 230000 mg/kg in dry season and 130000 - 167000 mg/kg in wet season (Table 5). Based on Figure 2 (b), the mean concentration of Al was higher during dry season as Al is the third most abundant element in earth's crust after Si 30, thus the both elements were found in high concentrations in the soil samples.

Ca is commonly found in soil in abundance. In this study, the concentration of Ca found in soil during dry and wet seasons ranged from 1550.00 - 2370.00 mg/kg and 1010.00 - 2810.00 mg/kg, respectively. Figure 3(a) shows that the mean concentration of Ca was higher during dry season (2012.50 mg/kg) compared to wet season (1915.00 mg/kg). Apart from that, K element found in soil samples was in the range of 10600.00 - 22000.00 mg/kg in dry season and 5930.00 - 19400.00 mg/kg in wet season. As presented in Figure 3(b), the mean concentration of K was also higher during dry season (16825.00 mg/kg) compared to wet season (12095.00 mg/kg).

The mean values for concentrations of Ti, Mn, Cl, Zr, and Cr during dry and wet seasons are illustrated in Figures 4 – 6, respectively. The mean values for concentrations of Ti, Mn, Cl, Zr and Cr during dry season were 13120.00 mg/kg, 505.75 mg/kg, 494.00 mg/kg, 4777.50 mg/kg, and 341.25 mg/kg, respectively. As for wet season, the mean values for concentration of the selected elements were as follow: Ti (9562.50 mg/kg), Mn (437.50 mg/kg), Cl (1274.25 mg/kg), Zr (6505.00 mg/kg), and Cr (279.00 mg/kg).

Based on Figure 7(a), the average concentration of Zn was higher during dry season (190.25 mg/kg) compared to wet season (150.25 mg/kg). According to Najib et al ^{31,} the sources of Zn in soil may derive from motor vehicle emissions. Among the metals studied, Zn had the lowest mean concentration during wet season. Figure 7(b) indicates higher Pb metal accumulated in soil during dry season (149.00 mg/kg) compared to wet season (152.75 mg/kg). Similar to Zn, Pb had the lowest mean

concentration during dry season than other studied metals. Based on the previous study, the Pb discharged from engine oil, gasoline, and used container carried by runoff in the sampling sites may contribute to the higher concentration of Pb in soil during the wet season ³². Even the EF value of Pb indicated it had moderately severe enrichment, although the amount of Pb in the soil samples in the study area was still in the range of unpolluted to moderately-polluted, as referred to its I-geo data in Table 7. Figure 8 shows that the mean concentration of Sn was higher during wet season (186.25 mg/kg) compared to dry season (161.75 mg/kg).

3.3 Pollution Indices

a) Enrichment Factor (EF)

The data of enrichment factor was used in order to determine the geochemical trends between all the sampling points. The mean EF values for each selected metal during dry and wet seasons are presented in Table 6 and Figure 9. As mentioned by Zhang and Liu ³³, if the value of EF > 1.5, the metal is derived from non-crustal materials - anthropogenic source. This is also supported by Kamaruzzaman et al ³⁴, who stated that EF value close to 1 represents crustal origin, while EF value more than 10 reflects non-crustal source.

As for dry season, the mean values of EF were observed to exceed the level of very severe enrichment as the EF values were above 50. Sn with the highest mean EF value of 39.07 indicated very severe enrichment in soil. Ca was found to have the lowest mean EF value less than 1, thus indicating deficiency to minimal enrichment during the seasons studied. However, K, Mn, and Cl showed no enrichment in soil, whereas Si, Al, Ti, Cr, and Zn were enriched slightly. However, EF of Pb reflected that Pb in the soil was in moderately severe enrichment and Zr was in severe enrichment.

The mean EF values for all metals in wet season were categorised between no enrichment and extremely severe enrichment (Table 6). Similarly, Sn (60.37) was detected to have the highest EF values during wet season. As Si EF was above 50, it showed that the soil was extremely severe enriched, followed by Zr (severe enrichment), Pb (moderately severe enrichment), Si, Al, Ti (minor enrichment), and Ca, K, Mn (no enrichment).

Overall, metals with EF values >1.5 in this study were Ti, Zr, Pb, and Sn from seasons and Si, Cl, Cr, and Zn for wet season. This probably derived from anthropogenic activities in this study area. Si and Ti were slightly enriched in the soil samples, yet abundant in soil. Sn may be released in dusts from windstorms, roads, and agricultural activities ³⁵. Deposition of Cl in soil mainly resulted from the application of fertiliser applications, rainwater, irrigation waters, dust, and air pollution ³⁶.

According to Liu et al. ³⁷, the accumulation of Cr, Pb, and Zn in the soil derives from farming practices, especially sewage irrigation. According to Rahman et al. ³⁸, the source of Zn is related to non-ferric metal industry and agricultural practice. Since the sampling sites were not in metal industry area, the anthropogenic source of Zn is suspected to derive from agricultural practices. Meanwhile, the sources of Pb in soil derived from the lead discharged from battery waste, engine oil, gasoline, and used container carried by runoff in the sampling sites ³².

b) Geo-accumulation Index (I-geo)

The I-geo that consists of seven levels ranging from unpolluted to very highly-polluted had been used to determine the degree of pollution in the soil sample. The I-geo values of each metal for dry and wet seasons are shown in Table 7 and Figure 10. Therefore, the average I-Geo values and classes were summarised. Referring to the mean values of I-geo data shown in Table 7, the soils at the both rubber cultivation sites were in the range of unpolluted to moderately / highly polluted. Si was categorised as moderately polluted (class 2) in dry season and moderately to highly polluted (class 3) for wet season. Meanwhile, other metals in both seasons had 0 as the I-geo values, which signified unpolluted. In summary, the soil at the rubber plantation was highly contaminated by Si metal. According to Matichenkov and Bocharnikova³⁹, Si is a basic mineral formatting element and has a large surface area that is able to absorb water, phosphates, potassium, nitrogen, aluminium, and heavy metals. The traditional practice of collecting ashes from the backyard of houses that accumulated over a period of time from burning firewood, twigs, and trashes; then applying them to fields before the commencement of cropping supplies silicon indirectly ⁴⁰ contributed to this result. However, Si showed minor enrichment in the soil sample. As Si is the second most abundant element in Earth's crust, the high accumulation of Si in soil might derive from the natural rock weathering processes.

c) Pollution Load Index (PLI)

The PLI is an easy and comparative way to assess the degree of metal pollution that gives a summative indication of the overall level of heavy metal toxicity in a particular sample ⁴¹. Based on the data obtained in this study (Table 8), the mean PLI values indicate that the rubber cultivation site was unpolluted with the studied metals during dry and wet seasons, as the PLI values were less than 1 for all stations. Even though very high contamination was detected from Si to a low extent of Pb, the calculated PLI revealed that the soil was uncontaminated.

3.0 Conclusion

The present study indicated that the Si metal in rubber plantation soil was highly contaminated during dry and wet seasons, whereas Pb and Zn displayed the lowest concentrations in dry and wet seasons, respectively. Based on the EF calculation, Ti, Zr, Pb, Sn, Si, Cl, and Zn were significantly enriched; while for I-geo, the soil was polluted by Si for both seasons. This suggests that the soil from rubber plantation might be associated to anthropogenic activities. However, regular monitoring of the contents of metals should be made in future to avoid any heavy metal toxicity upon human beings and the environment.



(Source: Google Earth) Note: Station 1: Kampung Kulim Station 2: Kampung Gemang

Figure 1. Maps of Study Area



Figure 5. Mean Concentration of (a) Cl and (b) Zr

(a)

Zr

(b)













Table 1. GPS Coordinate of Sampling Poi	int
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Station	Latitude	Longitude
1	5°44'29.1114"N	101°51'28.5834"E
2	5°45'26.0742"N	101°51'53.6832"E

Table 2. Enrichment Factor in Relation to the Sediment Quality

EF Value	Classification
EF < 1	No enrichment
EF < 3	Minor enrichment
$\mathbf{EF} = \mathbf{3-5}$	Moderate enrichment
$\mathbf{EF} = \mathbf{5-10}$	Moderate severe enrichment
EF = 10-25	Severe enrichment
$\mathbf{EF} = 25\mathbf{-50}$	Very severe enrichment
EF > 50	Extremely severe enrichment

Table 3. Geo-Accumulation Index (I-geo) Classes in Relation to Soil Quality

I-Geo	I-Geo Class	Soil Quality
0-0	0	Unpolluted
0-1	1	Unpolluted to moderately polluted
1-2	2	Moderately polluted
2-3	3	Moderately to highly polluted
3-4	4	Highly polluted
4-5	5	Highly to very highly polluted
5-6	>5	Very highly polluted

Table 4. Soil pH, Moisture Content (MC) And Total Organic Matter (TOM) During Dry and Wet Season

		Dry Season		Wet Season				
Station	Ph	MC (g)	TOM (%)	pН	MC (g)	TOM (%)		
G1	6.6	30.12	0.0006	6.5	33.83	0.0005		
G2	6.2	29.66	0.0005	6.5	34.27	0.0005		
K1	6.7	30.20	0.0006	6.6	51.07	0.0005		
K2	6.6	20.20	0.0005	6.8	25.46	0.0003		
Mean	6.5	32.55	0.0005	6.6	36.16	0.0004		
Range	6.2 - 6.7	20.20 - 30.20	0.0005 - 0.0006	6.5 - 6.8	25.46 - 51.08	0.0003 - 0.0005		

Metals	Dry Season					Wet Season					
Concentration (mg/kg)	G1	G2	K1	K2	Mean	G1	G2	K1	K2	Mean	
Si	642000.00	631000.00	636000.00	761000.00	667500.00	732000.00	769000.00	726000.00	796000.00	755750.00	
Al	216000.00	214000.00	230000.00	152000.00	203000.00	157000.00	130000.00	167000.00	130000.00	146000.00	
Ca	1950.00	2370.00	2180.00	1550.00	2012.50	2810.00	2050.00	1790.00	1010.00	1915.00	
K	13800.00	22000.00	10600.00	20900.00	16825.00	6450.00	19400.00	5930.00	16600.00	12095.00	
Ti	14900.00	13800.00	14700.00	9080.00	13120.00	10500.00	8470.00	11300.00	7980.00	9562.50	
Mn	487.00	602.00	583.00	351.00	505.75	404.00	467.00	548.00	331.00	437.50	
Cl	834.00	141.00	120.00	881.00	494.00	1070.00	2110.00	677.00	1240.00	1274.25	
Zr	5140.00	4830.00	4050.00	5090.00	4777.50	7240.00	6570.00	6220.00	5990.00	6505.00	
Cr	395.00	282.00	358.00	330.00	341.25	308.00	278.00	298.00	232.00	279.00	
Zn	281.00	127.00	199.00	154.00	190.25	135.00	170.00	161.00	135.00	150.25	
Pb	140.00	153.00	150.00	153.00	149.00	155.00	156.00	148.00	152.00	152.75	
Sn	172.00	164.00	150.00	161.00	161.75	189.00	181.00	199.00	176.00	186.25	

Table 6. Enrichment Factor(EF) of each Station During Dry and Wet Season

Elements	Dry Season					Wet Season				
	G1	G2	K1	K2	Mean	G1	G2	K1	K2	Mean
Si	0.9350	0.9279	1.0095	2.4496	1.3305	1.4562	1.9129	1.4938	3.1339	1.9992
Al	1.1381	1.1386	1.3209	1.7702	1.3420	1.1301	1.1700	1.2433	1.8518	1.3488
Ca	0.0212	0.0261	0.0259	0.0373	0.0276	0.0418	0.0381	0.0276	0.0297	0.0343
K	0.2705	0.4354	0.2264	0.9054	0.4594	0.1727	0.6495	0.1642	0.8795	0.4665
Ti	1.5584	1.4575	1.6758	2.0991	1.6977	1.5002	1.5132	1.6699	2.2564	1.7349
Mn	0.2853	0.3561	0.3722	0.4545	0.3670	0.3233	0.4673	0.4535	0.5242	0.4421
Cl	0.7411	0.1265	0.1162	1.7303	0.6785	1.2988	3.2026	0.8500	2.9788	2.0826
Zr	10.6197	10.0771	9.1203	23.2445	13.2654	20.4341	23.1865	18.1573	33.4572	23.8088
Cr	1.3148	0.9479	1.2989	2.4280	1.4974	1.4005	1.5807	1.4015	2.0877	1.6176
Zn	1.8132	0.8275	1.3996	2.1964	1.5592	1.1900	1.8737	1.4678	2.3549	1.7216
Pb	3.9675	4.3784	4.6332	9.5836	5.6407	6.0004	7.5514	5.9259	11.6450	7.7807
Sn	31.3651	30.1995	29.8137	64.8927	39.0678	47.0811	56.3790	51.2721	86.7648	60.3743

Table 7. I-geo Value and Classes for Both Season

	Dry S	eason	Wet Season		
Elements					
	I-geo Value	I-geo Class	I-geo Value	I-geo Class	
Si	2	2	3	3	
Al	0	0	0	0	
Ca	0	0	0	0	
K	0	0	0	0	
Ti	0	0	0	0	
Mn	0	0	0	0	
Cl	0	0	0	0	
Zr	0	0	0	0	
Cr	0	0	0	0	
Zn	0	0	0	0	
Pb	0	0	0	0	
Sn	0	0	0	0	

	Dry Season					Wet Season				
Element	G1	G2	K1	K2	Mean	G1	G2	K1	K2	Mean
PLI	0.0131	0.0101	0.0151	0.0214	0.0149	0.0132	0.0140	0.0133	0.0180	0.0146

Table 8. Pollution Load Index, (PLI) Values Observed during Dry and Wet Season

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