

GEOSTATISTICAL ASSESSMENT OF HEAVY METALS AND NUTRIENTS AVAILABILITY IN SOIL OF OIL PALM PLANTATION AFFECTED BY BAUXITE MINING

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ABSTRACT

This study describes the contamination of heavy metals (Cu, Zn, Mn, Pb and Fe) and their effect on K, Ca and Mg availability in oil palm cultivated areas affected by bauxite mining activities using the combinations of geostatistic and geospatial analysis. A total of 64 soil samples were collected covering a total area of 420.21 ha by grid sampling technique. Spatial distributions of the heavy metals were determined using semivariogram and mapped using ArcGIS. The mean concentrations of Cu ($138.94 \pm 79.08 \text{ mg kg}^{-1}$), Zn ($233.55 \pm 79.16 \text{ mg kg}^{-1}$), Mn ($847.88 \pm 267.02 \text{ mg kg}^{-1}$) and Fe ($249\ 703.71 \pm 101\ 408.72 \text{ mg kg}^{-1}$) in this study were greater than the background values, the 95% 'Investigation Levels' determined for Malaysia soil and Dutch target values. Geoaccumulation index showed that the contamination was in the order of $\text{Fe} > \text{Cu} > \text{Pb} > \text{Zn} > \text{Mn}$. Semivariogram analysis of pH, Mn, Zn and Fe was aligned with the principal component analysis results, showing the contamination source originated from a similar identical source. In correlation to the nutrients, only K_{ex} was found to be affected by the contaminants. These results provide a useful basis for the related agencies in identifying hotspots for future rehabilitation programs.

Keywords: geoaccumulation index, geospatial, GIS, PCA

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INTRODUCTION

The palm oil industry is a prominent contributor to Malaysia's economy. In 2020, oil palm planted area in Malaysia was approximately about 5.87 million hectares in Peninsular Malaysia covering about 2.74 million hectares, while Sabah and Sarawak were 1.54 million hectares and 1.55 million hectares, respectively (MPOC, 2020). After Johor, Pahang had the second-largest oil palm plantation (732 052 ha).

In 2014, bauxite mining activities in Pahang received a lot of public attention. A total of 18 000 ha of land in Kuantan had been extensively used for bauxite mining purposes including the area of oil palm plantations. The mining operation has been reported to cause land subsidence, which destroyed the soil aggregation and caused environmental damages such as loss of topsoil and a decline in soil fertility due to erosion (Amanah and Yunanto, 2021; Prematuri *et al.*, 2020). During the mining process, about 20 m of soil surface was stripped to obtain bauxite ores, which caused the soil to lose its crucial topsoil layer. This inevitably resulted in the declining amount of organic layers in the soil, which is the integral site of nutrient storage and exchange. The loss of this layer will affect nutrient absorption and their availability to the crops, which ultimately results in declining yield production (Zhang *et al.*, 2020). Additionally, mining operations also alter soil chemical properties and pH values. Changes in pH will affect the availability of soil heavy metals

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(HMs) and cause toxicity symptoms to the oil palm trees. Studies conducted by Kusun *et al.* (2016) reported that sediments affected by bauxite mining activities in Kuantan had a higher content of As in a few localities with a value of about 8 mg kg^{-1} , where the source of contamination was identified to be from the mining exploration that mobilised the As. Analysis of geoaccumulation index (I_{geo}) classified the As as slightly polluted in the study area. Other HMs were within the permissible values. However, slightly higher amounts of Mg, Na, Al, Fe, As and Pb was observed in the water bodies nearby. The study stated that anthropogenic sources such as fertilisers and chemical industry effluent could be the sources of the contamination. This can also be influenced by open-cast bauxite mining, which caused the soil surface to be eroded and carried to adjacent rivers.

Geostatistics and geographic information system (GIS) techniques have been widely employed to quantify the spatial distribution of soil properties to reduce uncertainties, minimising costs and to identify pollution sources, especially in areas of mined soil (Chen *et al.*, 2021; Kulikova *et al.*, 2019). It has additional advantages in research as its analysis includes the characteristics of space and time changes. Through these, researchers can effectively determine the relationship between HMs indicators and their potential sources of pollution (Shen *et al.*, 2021; Xu *et al.*, 2021; Zhang and Yang, 2017). Research related to HMs contamination in oil palm plantations impacted by bauxite mining is crucial. A visit to the study site revealed that the mined region was left abandoned without any rehabilitation efforts. Thus, preliminary data on soil quality is needed to assess the impact of bauxite mining on the surrounding oil palm plantation areas. The outcome of this study can be used for future rehabilitation and oil palm replanting program. With the aid of geospatial technologies, researchers are able to simplify the process of interpreting data in quantifying variability in the field. The aims of this research were: (a) to determine the contents of selected soil HMs, which are Cu, Zn, Mn, Pb and Fe in bauxite mining and oil palm cultivated area and their effect on selected macronutrient availability, (b) to evaluate the contamination of soil HMs (Cu, Zn, Mn, Pb and Fe) using multivariate statistical analysis and environmental index, and (c) to quantify and generate a spatial map of HMs (Cu, Zn, Mn, Pb and Fe) using geostatistical and geospatial analysis.

MATERIALS AND METHODS

Study Area

The study was conducted in Bukit Goh, Kuantan, Pahang, located between latitude $3^{\circ}55'30''$ to $3^{\circ}55'0''$ N and longitude $103^{\circ}16'0''$ to $103^{\circ}17'0''$ E,

covering 420.21 ha (Figure 1). Bauxite mining and oil palm cultivation activities covered approximately 204.88 ha and 215.33 ha of the study area, respectively. The soil was categorised into Kuantan soil series, which is very fine, oxidic, isohyperthermic and brown Tipik Akrolemoks. The drainage was slightly excessive with deep soil profile ($>2 \text{ m}$ depth) and had a gently undulating terrain (4%-20% or 2-10° slopes) over quaternary basalts. The annual rainfall was about $2400 \text{ mm year}^{-1}$ with an average temperature of 28°C .

Soil Sampling and Analysis

Soil sampling was performed using systematic sampling technique based on a regular grid size of $300 \times 300 \text{ m}$ within the study area. A total of 64 soil samples were collected using a corer at depth of 0-20 cm. Each sampling position was recorded using global positioning system (GPS) (GPSMAP 64s, Garmin, U.S.A).

Soil samples were air-dried, ground and subsequently sieved ($<2.00 \text{ mm}$). Soil pH was determined using potassium chloride (KCl) with 1:2.5 ratio. Total organic carbon (TOC) was determined using Walkley-Black dichromate methods (Walkley Black, 1934). Soil exchangeable K, Ca and Mg were extracted using Mehlich 3 procedures (Mehlich, 1984), which is suitable for acidic soil type. Total content of Cu, Zn, Mn, Pb and Fe were digested using mixed acid with ratio of 3: 3.5: 3.5; HF: HNO_3 : HCl and analysed by using inductively coupled plasma mass spectrometry (ICP-MS) (Elan 9000, Perkin Elmer, U.S.A). For quality and precision assurance of the analysis, reference material (SRM 1694a) of National Institute of Standards and Technology (NIST) was used. The percentage of recovery for the metals studied ranged between 85% and 120%. Duplicate samples and blanks for each sample were performed to ensure the precision and accuracy of the analysis.

Statistical Analysis

All statistical analyses were performed using SPSS package version 26.0 and were tested using normality test of Shapiro-Wilk. As the data were of the compositional geological component, non-normal distribution data were logarithm transformed. The degree of soil variability in the study area, was evaluated through the coefficient of variation (CV) value. Ranking of the CV were categorised into three classes as in Table 1 (Wilding, 1985).

Geostatistical Analysis based on GIS

The HMs spatial interpolation was done using ArcGIS v.10.8. The normality of soil HMs concentrations in this study was assessed through the Kolmogorov-Smirnov (K-S) test and non-normal data were transformed using the log transformation

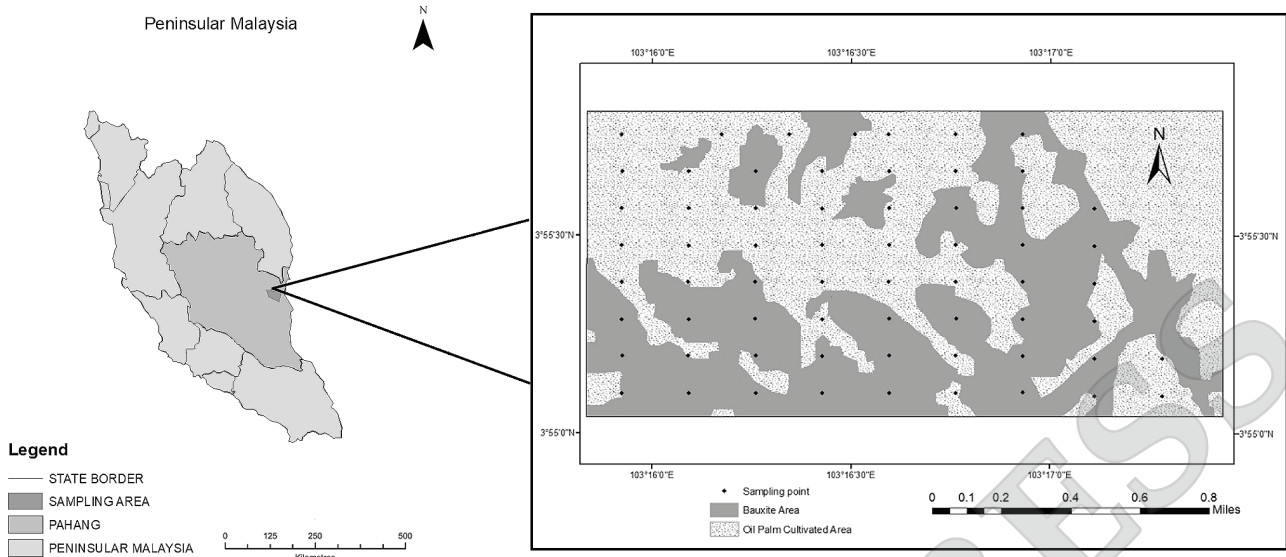


Figure 1. Location of study area in Kuantan, Pahang, Malaysia and the soil sampling points.

TABLE 1. RANGE OF COEFFICIENT OF VARIATION

CV (%)	Class
$X < 15$	Low variability
$15 \leq X \leq 35$	Moderate variability
$X > 35$	High variability

Source: Wilding (1985).

method. Experimental semivariogram was fitted using theoretical models; exponential, linear, spherical and gaussian models. A semivariogram is expressed using the regionalised variable theory and intrinsic hypotheses (Nielsen and Wendroth, 2003) as Equation (1):

$$\gamma(h) = \frac{1}{2N(h)} \sum_{i=1}^{N(h)} [Z(x_i) - Z(x_i + h)]^2 \quad (1)$$

where $Z(x_i)$ is the value of the variable Z at location of x_i , h the lag and $N(h)$ the number of pairs of sample points separated by h . Theoretical semivariograms were used to fit the empirical semivariogram models to generate geostatistical parameters, such as structured variance (C_1), nugget variance (C_0), and sill variance ($C_0 + C_1$) and distance parameter (h). The fitted model was adopted based on high R^2 ($> 50\%$) and low residual sum square (RSS) (Wang *et al.*, 2021). Ordinary kriging interpolation was employed after semivariogram models were constructed.

Geoaccumulation Index (I_{geo})

Geoaccumulation Index (I_{geo}) for HMs assessment was developed according to the world wide standard

shale values (Muller, 1969). This index is denoted as Equation (2):

$$I_{geo} = \log_2 \left[\frac{C_n}{1.5 B_n} \right] \quad (2)$$

where C_n is the measured concentration of the element in soil, B_n is the value of geochemical background and the fix 1.5 value used to investigate natural changes in the level of a certain chemical in the environment and to identify extremely minor anthropogenic influences. Muller (1969) classified and described the Geoaccumulation Index (I_{geo}) into classes (Table 2).

RESULTS AND DISCUSSION

Statistical Analysis

Table 3 shows the descriptive statistics of soil pH and contents of selected HMs examined. The HMs in soil of the study areas were compared and referred to the natural background values published by Wedepohl (1995), Dutch target and intervention values for soil remediation (Dutch Target and Intervention Values, 2000), which has been widely used in HMs studies (Rudzi *et al.*, 2018) and the 95% 'Investigation Levels' determined for Malaysia soil (Zarcinas *et al.*, 2004).

The soil pH of the study area ranged between 3.65 to 4.82, indicating acidic conditions. This is similar to the pH of post mining soil reported by Prematuri *et al.* (2020), with value of 4.52 ± 0.09 . Soil pH is one of the most important elements that controls cation mobility and governs HMs solubility in soil (Li *et al.*,

TABLE 2. I_{geo} CLASSES

Class	Value	Soil quality
0	$I_{geo} \leq 0$	Uncontaminated
1	$0 < I_{geo} \leq 1$	Uncontaminated to moderately contaminated
2	$1 < I_{geo} \leq 2$	Moderately contaminated
3	$2 < I_{geo} \leq 3$	Moderately to heavily contaminated
4	$3 < I_{geo} \leq 4$	Heavily contaminated
5	$4 < I_{geo} \leq 5$	Heavily to extremely contaminated
6	$I_{geo} \geq 5$	Extremely contaminated

availability in the environment is influenced by anthropogenic activities, while CV less than 15% may only be occurred by the natural process such as geological and weathering factors. From the results, only soil pH showed a low variability (CV<15%), which indicated its changes within the study area were caused by natural factors such as climate, weathering process, topography and geological factors. While, Zn and Mn showed a moderate variability, with values of 33.63% and 31.49%, respectively, Cu, Pb and Fe showed a higher CV. This suggested that anthropogenic sources, such as

TABLE 3. SUMMARY STATISTICS OF SELECTED SOIL PROPERTIES AND HEAVY METALS CONTENT IN SOIL OF THE STUDY AREA (n=64)

Item	Range	Mean \pm SD	CV (%)	Skewness	Kurtosis	Other studies references		
						a	b	c
pH	3.65 - 4.82	4.18 \pm 0.28	6.59	0.02100	-0.91			
TOC	0 - 1.80	0.43 \pm 0.33	76.19	1.63000	4.36			
Cu (mg kg ⁻¹)	22.86 - 315.37	138.94 \pm 79.08	56.47	0.38000	-0.98	25.0	50	36
Zn (mg kg ⁻¹)	74.82 - 377.37	233.55 \pm 79.16	33.63	-0.20000	-0.99	65.0	95	140
Mn (mg kg ⁻¹)	348.37 - 1425.11	847.88 \pm 267.02	31.49	-0.00016	-0.32	716.0	n.a	n.a
Pb (mg kg ⁻¹)	17.60 - 171.06	57.58 \pm 23.91	41.19	1.66000	6.87	14.8	65	85
Fe (mg kg ⁻¹)	23 459.12 - 446 985.00	249 703.71 \pm 101 408.72	40.29	-0.25000	-0.66	43200.0	n.a	n.a

Source: a) Continental crust value (Wedepohl *et al.*, 1995), b) The 95% 'Investigation Levels' determined for Malaysia soil (Zarcinas *et al.*, 2004), and c) Dutch Target and Intervention Values for soil remediation (Dutch Target and Intervention Values, 2000).

2022). Lower value of soil pH in oil palm plantation is common due to the regular application of chemical and ammonium based fertiliser, high decomposition of organic materials, nutrients uptake by oil palm and other agricultural practices. Although oil palm is relatively tolerant to acidic soil, attention should be given as soil with low pH could result in unbalance nutrient and further could cause toxicity effect. At low soil pH, the availability of acidic cations such as Fe, Cu, Zn, Mn will be higher at the CEC site and this will suppress the availability of K, Ca and Mg.

The content of Cu, Zn, Mn, Pb and Fe were greater than the background values (Wedepohl, 1995). This indicated that the soil of the bauxite mining and oil palm cultivated area could be possibly contaminated with Cu, Zn, Mn, Pb and Fe. However, in comparison to the Dutch target and intervention values for soil remediation and the 95% 'Investigation Levels' determined for Malaysia soil, the mean content of Pb obtained in this study was found to be lower. The content of Cu and Zn was two to three times higher than the Dutch target value and the 95% 'Investigation Levels' determined for Malaysia soil.

The coefficient of variation (CV) value aids in determining the HMs variability (Table 1). For HMs with CV greater than 35%, it is suggested that their

mining and agricultural practises including the use of fertilisers, manures, pesticides, and the removal of the top soil layer could be the cause of Cu, Pb, and Fe accumulation.

Analysis of Pearson Correlation

For analysis of Pearson correlation, the data were logarithm transformed and fitted to normality. The correlation analysis showed that Cu, Zn and Mn had a significant positive correlation with Fe at $p < 0.01$ of 0.82, 0.63 and 0.75, respectively (Table 4) suggesting the possibility of their common origin. Abundant amount of this HMs might be bound together with Fe in the parent material of the soil as it is the major component of oxisols soil. The significant positive correlation between the HMs explained that the contamination of the HMs in the soil could be possibly from similar sources of contamination (Doabi *et al.*, 2019). The correlation coefficients between Cu-Zn, Cu-Mn, Cu-Fe, Zn-Mn, Zn-Fe and Mn-Fe were relatively larger than 0.5, explaining a strong correlation among the HMs, indicating a homologous sources of contamination (Wang *et al.*, 2021). However, Pb did not show any significant correlation with the HMs. It can be hypothesised that Pb could be primarily

TABLE 4. PEARSON'S CORRELATION COEFFICIENT OF THE SOIL HMs (n=64)

		Correlation matrix ^a					
	pH	TOC	Cu	Zn	Mn	Pb	Fe
pH	1.000	-.276	.236	.226	.149	.189	.207
TOC		1.000	-.166	-.189	-.214	-.185	-.162
Cu			1.000	.849*	.808*	.200	.815*
Zn				1.000	.754*	.462	.627*
Mn					1.000	.263	.745*
Pb						1.000	-.045
Fe							1.000

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

generated from lithogenic or other point sources of contamination.

Further analysis by using PCA found that two eigenvalues were obtained (F1 and F2), with a total variance of 68.29% (Table 5). The first factor (F1) had 49.80% of the total variance and was significantly loaded with Cu, Zn, Mn and Fe. Copper had the most significant loads with value of 0.94 followed by Fe, Mn and Zn. The same trend had been initially observed during the correlation analysis, where a significant relationship was found among Zn, Mn, Fe with Cu. Significant correlation obtained among F1 indicated their homologous origin, probably from anthropogenic sources, notably from the mining activities, long term agricultural practices, applications of chemical fertilisers and acceleration of soil geochemical weathering that could be caused by the long-term mining exposure. The second factor, F2, was dominated by pH and Pb, which accounts 18.50% of the total variance. The sources of contamination in the study area could be classified into two categories based on the results obtained.

TABLE 5. ROTATED COMPONENT MATRIX FOR THE SOIL HMs (n=64)

Element	Component	
	1	2
pH	0.194	0.575
TOC	-0.169	-0.169
Cu	0.944	0.091
Zn	0.805	0.365
Mn	0.883	0.154
Pb	-0.069	0.788
Fe	0.920	0.014
Eigenvalue	3.487	1.293
Variance contribution rate (%)	49.813	18.472
Cumulative variance contribution rate (%)	49.813	68.285

Geostatistic and Semivariogram Analysis

Semivariogram analysis showed that pH, Cu and Fe fit to the spherical model, while Mn and Pb fit to the exponential model (Table 6). Whereas Zn fits to the Gaussian model and lastly, TOC fits to the linear model. The nugget variance ranged from 1.00×10^{-5} to 0.19. The obtained values were less than 30% of the sill value, indicating good spatial continuity and minimal measurement errors (Paz-Gonzalez *et al.*, 2001).

Range value indicates the distance by which the HMs content change. The range obtained from the semivariogram analysis varied between 119 to 1262 m, with the lowest being in Pb followed by Zn, Mn, pH, Cu, Fe and TOC. The TOC had a longer effective range (1262 m) indicating that TOC had a better spatial structure and less variation caused by extrinsic factors. The soil range for pH, TOC, Cu and Fe exceeded the distance of 300 m between points. Nevertheless, Zn, Mn and Pb had range value less than 300 m, suggesting a closer sampling procedure should be done in future.

A quotient level of spatial distribution (X) in this sample was categorised into three different levels; $< 25\%$ resembles strong spatial dependence, $25\% > X > 75\%$ resembles moderate spatial dependence and $X > 75\%$ resembles weak spatial dependence (Cambardella *et al.*, 1994). Nugget to sill ratio indicates whether the geographical distribution of HMs is due to intrinsic or external reasons (Guan *et al.*, 2017). The soil pH, Mn, Zn, Pb and Fe were observed to have strong spatial autocorrelations indicating that natural factors were responsible for their distributions in the studied area. However, TOC and Cu showed a moderate spatial dependence reflecting their distributions could be from the mixed inputs of natural and anthropogenic sources. The semivariogram analysis revealed that Mn, Zn and Fe were grouped together in the same group as PCA, indicating identical contamination causes. However, pH, Pb, and Cu had a contradictory results with the PCA. This could be due to the robust estimation of semivariogram, which is more extensive than PCA as it considers each data's coordinates (geographically). Semivariogram calculates and compares data points in all directions. Numerous internal factors such as pH, organic matter content, texture, redox state influenced the HMs dynamics in soil. These soil characteristics are thought to be a significant source of variation for HMs. Discussing about HMs distributions, the variation existed in the study area could be due to two soil-forming factors; the geological nature of the rock and topographic influences. In a small scale and stable natural landscape, such as in the study area, the parent material can be said to be homogenous, suggesting that topography could be the only source of variation. The studied area is characterised by a

TABLE 6. BEST FITTED SEMIVARIANCE MODELS AND THEIR PARAMETERS

Parameter	Nugget variance C_0	Structural sill (C_0+C)	Range A (m)	Nugget to sill ratio $C_0/(C_0+C)$ (%)	R^2	RSS	Model variogram model type
pH	1.0×10^{-5}	4.6×10^{-3}	613.00	0.22	0.95	3.145×10^{-7}	Spherical
TOC	0.0810	0.12	1262.91	68.00	0.52	6.08×10^{-4}	Linear
Cu	0.1900	0.45	903.00	42.00	0.98	5.43×10^{-4}	Spherical
Mn	0.0090	0.12	291.00	7.50	0.74	8.25×10^{-5}	Exponential
Zn	0.0120	0.15	167.00	8.00	0.87	2.21×10^{-4}	Gaussian
Pb	0.0085	0.16	119.00	5.30	0.77	2.70×10^{-5}	Exponential
Fe	0.0010	0.33	1 160.00	0.30	0.97	1.75×10^{-3}	Spherical

hilly topography with moderate slope. Irregular terrain and various activities conducted such as mining and long-term agricultural practices, may all be the factors in the circumstance, where soil is exposed to activities that are made by humans. Natural geochemical processes such as weathering can be accelerated through human anthropogenic activities by the changes and disturbance in soil chemical conditions such as pH, organic matter content and redox state. This will allow the stable mineral to become soluble and mobile in the environment. Therefore, we assume that in this study, soil forming factors, parent material, topography and human factors may influence the spatial variability of HMs.

Kriging Interpolation

The spatial distributions of pH, TOC, Cu, Zn, Mn, Pb and Fe are visualised in Figure 2(a - g). The soil of the study area was classified as acidic, ranging from 3.60 to 4.82. Low soil pH in the study area is expected to cause adverse effect to the oil palm's growth and development. It also affect the availability of soil K, Ca and Mg as the cation exchange site will be dominated by the ion H^+ . In addition, it will also increase the toxicity and mobility of HMs (Olafisoye *et al.*, 2020).

The distribution of TOC showed that organic carbon were found low throughout the studied region. This could be caused by the removal of the top soil layers for the mining purposes (Lee *et al.*, 2017). The lower TOC content indicates insufficient organic matter in the soil. Organic matter in soil is essential as it creates a good soil structure and promoted microbial activities. By enhancing the aggregate stability, the soil water infiltration, holding capacity and aeration will be improved. Contrasted to soil with poor porosity and high bulk density value, the growth of the crops such as oil palm will be impeded. In addition, poor soil water holding capacity and infiltration will also eventually cause mud floods to the plantation area and this will

inevitably impede the oil palm growth and lead to a lower yield.

A total of 92.5% of the study area had elevated levels of Cu (Figure 2c). Only about 18.0% of area in Figure 2c showed a non-contaminated region, with Cu lower than 36 mg kg^{-1} . Most of agricultural soil contaminated by HMs were reported to be associated with the application of phosphate fertilisers (Wong *et al.*, 2017). Although the levels of trace elements in the fertilisers were within permissible limits, the accumulation of leached residues is still a worry. Copper-based fungicides and pesticides also are the main source of Cu contamination in agricultural soil (Apori *et al.*, 2018). Copper resides on the surface of plant tissues and can be washed off resulting to a higher Cu content in soil.

The distributions of Zn showed that almost 90.0% of the study area had Zn higher than 140 mg kg^{-1} . According to Ismail *et al.* (2018), Zn is naturally abundant in sedimentary rocks and this may be the reason for the higher distribution of Zn observed. The region with higher Zn level was in the area of bauxite mining rather than the oil palm cultivated area. This could be the consequence of the extensive mining operations that exposed the soil without cover crops. Once the land was left barren, the soil was exposed to an extensive weathering process. This leads to modification of soil chemical properties and resulted in availability of HMs to the environment.

Manganese was found higher at the northern and central part of the study area (Figure 2e). More than 70% of the study area had Mn higher than 716 mg kg^{-1} . Manganese is also a component of the silicate mineral structure and one of the most common and widely distributed metals in nature. It can be commonly found in the structure of rocks and soils (Ismail *et al.*, 2018). Nevertheless, the source of Mn might also be from the Mn-containing fertilisers used in the oil palm cultivation area. Low soil pH will intensify Mn toxicity and will cause necrotic lesion to the oil palm fronds and restrict nutrients availability.

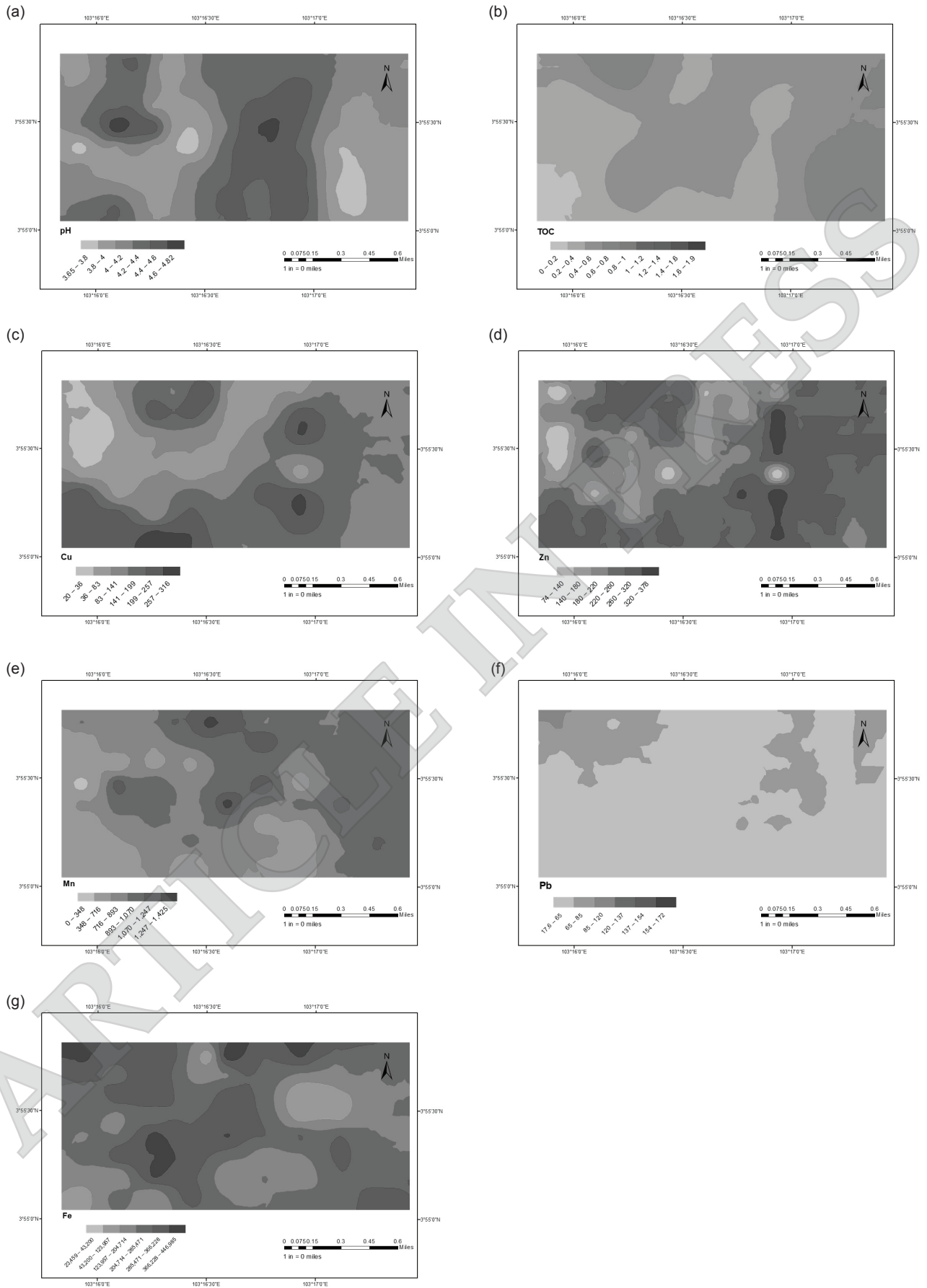


Figure 2. Spatial distribution of heavy metals in soil (a) pH, (b) TOC, (c) Cu, (d) Zn, (e) Mn, (f) Pb and (g) Fe.

Almost 100% of the study area (Figure 2f) had lower content of Pb compared to the Dutch target and intervention values for soil remediation. Sources of Pb in the soil was not known specifically. However, its availability can be from the exhaust of automobiles, fertilisers and pesticides (Yang *et al.*, 2018).

A total of 100% of the study region had Fe greater than the background values. Soil of Kuantan series is composed of goethite (FeOOH) and hematite (Fe₂O₃), where the main component of their silicate mineral is the Fe element. Optimum amount of Fe in soil will enhance the nutrients uptakes by the plant. However, elevated level of Fe will cause toxicity effect and restrict the oil palm development (Nurmalasari *et al.*, 2016). Iron contamination has the ability to alter the plant's morphology, anatomy, and physiology. To reduce the toxicity occurrence, it is crucial to maintain the pH around neutral conditions and avoid pH lesser than 4.5.

The spatial distribution maps depict the study area with several zones of HMs concentration. With the aid of the map, nutrient site-specific management program and rehabilitation strategy can be developed. In addition, the contaminated area also can be measured quantitatively and this allows the government and related agencies to conduct a cost-effective analysis for a land restoration programme based on the identified area.

Macronutrients Availability

Oil palm yield and growth are significantly influenced by soil nutrients. Table 7 shows the content of available nutrients; K_{ex}, Ca_{ex} and Mg_{ex}. The mean values of K_{ex} and Mg_{ex} were compared to the values suggested for oil palm published by Goh (1977). The mean of K_{ex} was slightly lower compared to Ca_{ex} and Mg_{ex}. This could be caused by the higher contents of Ca_{ex} and Mg_{ex} observed throughout the study area. Elevated level of Ca_{ex} and Mg_{ex} reduced the possibility of K_{ex} being attached to the cation exchange site as Ca and Mg had a higher affinity attached to the CEC site.

The CV values for K_{ex}, Ca_{ex} and Mg_{ex} were more than 35%, denoting high variability. This could be caused by the application of fertilisers especially in the oil palm cultivated area. Factors of uneven topography and human activities caused the cations to be easily leached and mobile throughout the study area.

The K_{ex}, Ca_{ex} and Mg_{ex} shows a positive but low coefficient value with TOC ($p < 0.05$) (Table 8). The K_{ex}, which was found low showed a negative correlation with Cu, Zn, Mn, Pb and Fe. This could be the effect of higher Cu, Zn, Mn, and Fe dominating the soil's cation exchange site. It led to the lower availability of soil to retain K⁺. In addition, K⁺ in soil has lower affinity for cation adsorption relative to Ca²⁺ and Mg²⁺, which might result in lower K⁺ value in the soil.

Geoaccumulation Index (I_{geo})

Figure 3 displays the results of the I_{geo} in the study area. The contamination was in the order of Fe > Cu > Pb > Zn > Mn. The Mn was categorised as "uncontaminated" as the I_{geo} value was less than 0.

The Cu, Zn, Pb and Fe of the soil in the study region were categorised into moderately contaminated. Iron showed the highest I_{geo} value with an average of 1.77 ± 0.80. It showed a consistent trend as initially high Fe was observed compared to the reference value. However, although Pb showed a lower distribution throughout the region, the I_{geo} value was 1.26 ± 0.59 reflecting a moderate contamination within the region.

CONCLUSION

The mean concentrations of Cu, Zn, Mn and Fe in the study area were found greater than the background

Geoaccumulation Index Value (I_{geo}) for Cu, Zn, Mn, Pb and Fe

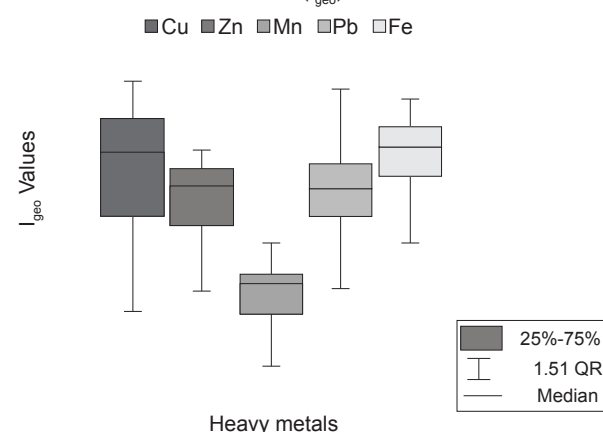


Figure 3. Boxplot of I_{geo} value for Cu, Zn, Mn, Pb and Fe with (n=64).

TABLE 7. DESCRIPTIVE STATISTICS OF SELECTED MACRONUTRIENTS IN SOIL OF THE STUDY AREA (n=64)

Nutrients	Mean ± STD (mg kg ⁻¹)	Range (mg kg ⁻¹)	Coefficient of variation (%)	Reference (Goh, 1977) (mg kg ⁻¹)		
				Very low	Low	Moderate
K _{ex}	70.57±45.49	0.55-190.37	64	<31.2	31.2-78.0	78.0-97.5
Ca _{ex}	37.34±41.57	0.00-149.26	110	n.a	n.a	
Mg _{ex}	35.81±18.72	3.05-139.75	52	<9.6	9.6-24.0	24.0-30.0

TABLE 8. PEARSON CORRELATION COEFFICIENTS OF HMS AND SELECTED MACRONUTRIENTS (n=64)

Nutrients	pH	TOC	Cu	Zn	Mn	Pb	Fe
K _{ex}	-0.08	0.19	-0.53	-0.44	-0.30	-0.02	-0.46
Ca _{ex}	-0.10	0.17	-0.45	-0.33	-0.34	0.06	-0.51
Mg _{ex}	-0.09	0.20	-0.47	-0.38	-0.39	0.05	-0.45

values, the 95% 'Investigation Levels' determined for Malaysia soil and Dutch target values. Analysis by I_{geo} showed that the contamination was in the order of Fe > Cu > Pb > Zn > Mn. Only K_{ex} was affected by the HMs contamination, while Ca_{ex} and Mg_{ex} showed a higher value and were not affected by the mining activities. The effectiveness of using multivariate statistical analysis and geostatistic in evaluating HMs concentration has been demonstrated in this study. It was identified that the availability of Mn, Zn, Cu, Fe and Pb in the study area could be from the natural component such as parent material and weathering processes. However, anthropogenic activities such as mining and agriculture practices had accelerated the HMs availability through soil properties changes and caused contamination. The geospatial mapping of HMs provides a good visualization in determining hotspot areas of each HMs. In future, government and related agencies will be able to utilise the HMs variability map for soil rehabilitation program and implementing site specific fertilizing scheme to enhance oil palm productivity.

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REFERENCES

- Amanah, F and Yunanto, T (2021). Oil palm plantation in ex-coal mined site: A case study in East Kalimantan, Indonesia. *IOP Conf. Ser. Earth Environ. Sci.*, 918(1): 012049. DOI: 10.1088/1755-1315/918/1/012049.
- Apori, O S; Hanyabui, E and Asiamah, Y J (2018). Remediation technology for copper contaminated soil: A review. *Asian Soil Res. J.*, 1(3): 1-7. DOI: 10.9734/ASRJ/2018/45322.
- Cambardella, C A; Moorman, T B; Novak, J M; Parkin, T B; Karlen, D L; Turco, R F and Konopka, A E (1994). Field-scale variability of soil properties in Central Iowa soils. *Soil Sci. Soc. Am. J.*, 58(5): 1501-1511. DOI: 10.2136/sssaj1994.03615995005800050033x.
- Chen, G; Yang, Y; Liu, X; Wang, M (2021). Spatial distribution characteristics of heavy metals in surface soil of Xilinguole coal mining area based on semivariogram. *ISPRS Int. J. Geo-Inf.*, 10: 290. DOI: 10.3390/ijgi10050290.
- Doabi, S A; Karami, M and Afyuni, M (2019). Heavy metal pollution assessment in agricultural soils of Kermanshah province, Iran. *Environ. Earth Sci.*, 78(3): 70. DOI: 10.1007/s10661-016-5225-3.
- Dutch Target and Intervention Values (the New Dutch List) (2000). Annexes - circular on target values and intervention values for soil remediation. https://www.esdat.net/environmental%20standards/dutch/annexes_i2000dutch%20environmental%20standards.pdf, accessed on 16 August 2020.
- Goh, K J (1977). Fertilizer recommendation systems for oil palm : Estimating the fertilizer rates. *Proc. of MOSTA Best Practices Workshops: Agronomy and Crop Management*. Malaysia. p. 235-268.
- Guan, F Y; Xia, M P; Tang, X L and Fan, S H (2017). Spatial variability of soil nitrogen, phosphorus and potassium contents in Moso bamboo forests in Yong'an City, China. *Catena*, 150: 161-172. DOI: 10.1016/j.ecolind.2020.107285.
- Ismail, S N S; Abidin, E Z; Praveena, S M; Rasdi, I; Mohamad, S and Ismail, W M I W (2018). Heavy metals in soil of the tropical climate bauxite mining area in Malaysia. *J. Phys. Sci.*, 29: 7-14. DOI: 10.21315/jps2018.29.s3.2.
- Kulikova, T; Hiller, E; Jurkovic, L; Filová, L; Šottník, P and Lacina, P (2019). Total mercury, chromium, nickel and other trace chemical element contents in soils at an old cinnabar mine site (Merník, Slovakia): Anthropogenic versus natural sources of soil contamination. *Environ. Monit. Assess.*, 191: 263. DOI: 10.1007/s10661-019-7391-6.
- Kusin, F M; Syazwan, M; Rahman, A and Madzin, Z (2016). The occurrence and potential ecological risk assessment of bauxite mine-impacted water and sediments in Kuantan, Pahang, Malaysia. *Environ. Sci. Pollut. Res.*, 24: 1306-1321. DOI: 10.1007/s11356-016-7814-7.

- Lee, K Y; Ho, L Y; Tan, K H; Tham, Y Y; Ling, S P; Qureshi, A M; Ponnudurai, T; Nordin, R and Cheah, J (2017). Environmental and occupational health impact of bauxite mining in Malaysia: A review. *Int. Med. J. Malays.*, 16(2): 137-150. DOI: 10.31436/imjm.v16i2.346.
- Li, Q; Wang, Y; Li, Y; Li, L; Tang, M; Hu, W; Li, C and Ai, S (2022). Speciation of heavy metals in soils and their immobilization at microscale interfaces among diverse soil components. *Sci. Total Environ.*, 825: 153862. DOI: 10.1016/j.scitotenv.2022.153862.
- MPOC (2020). Malaysian palm oil sector performance in 2020 and market opportunities. <https://mpoc.org.my/malaysian-palm-oil-sector-performance-in-2020-and-market-opportunities/>, accessed on 12 April 2022.
- Mehlich, A (1984). Mehlich 3 soil test extractant: A modification of Mehlich 2 extractant, communications in soil science and plant analysis. *Commun. Soil Sci. Plant Anal.*, 15: 1409-1416. DOI: 10.1080/00103628409367568.
- Muller, G (1969). Index of geoaccumulation in sediments of the Rhine River. *Geo J.*, 2(1): 108-118.
- Nielsen, D R and Wendroth, O (2003). Spatial and temporal statistics- sampling field soils and their vegetation. *GeoEcology Textbook*. Catena-Verlag, Reiskirchen Germany. 416 pp.
- Nurmalasari, A I; Putra, E T S and Yudono, P (2016). Root morphology of eight hybrid oil palms under iron (Fe) toxicity. *Ilmu Pertanian (Agric. Sc.)*, 1(1): 13-18. DOI: 10.22146/ipas.11254.
- Olafisoye, O B; Fatoki, O S; Oguntibeju, O O and Osibote, O A (2020). Accumulation and risk assessment of metals in palm oil cultivated on contaminated oil palm plantation soils. *Toxicol. Rep.*, 7: 324-334. DOI: 10.1016/j.toxrep.2020.01.016.
- Paz-Gonzalez, A; Taboada Castro, M T and Vieira, S R (2001). Geostatistical analysis of heavy metals in a one-hectare plot under natural vegetation in a serpentine area. *Can. J. Soil Sci.*, 81(4): 469-479. DOI: 10.4141/s00-095.
- Prematuri, R; Turjaman, M; Sato, T and Tawaraya, K (2020). Post bauxite mining land soil characteristics and its effects on the growth of *Falcataria moluccana* (Miq.) Barneby & J. W. Grims and *Albizia saman* (Jacq.) Merr. *Appl. Environ. Soil Sc.*, 5: 6764380.
- Rudzi, S; Ho, Y and Kharni, A I (2018). Heavy metals contamination in paddy soil and water and associated dermal health risk among farmers. *Malaysian J. Med. Health Sci.*, 14(SP2): 2-10.
- Shen, W; Hu, Y; Zhang, J; Zhao, F; Bian, P and Liu, Y (2021). Spatial distribution and human health risk assessment of soil heavy metals based on sequential Gaussian simulation and positive matrix factorization model: A case study in irrigation area of the Yellow River. *Ecotoxicol. Environ. Saf.*, 225: 112752. DOI: 10.1016/j.ecoenv.2021.112752.
- Walkley, A J and Black, I A (1934). Estimation of soil organic carbon by the chromic acid titration method. *Soil Sci.*, 37: 29-38.
- Wang, R; Zou, R; Liu, J; Liu, L and Hu, Y (2021). Spatial distribution of soil nutrients in farmland in a hilly region of the pearl river delta in China based on geostatistics and the inverse distance weighting method. *Agric. (Switz.)*, 11(1): 1-12. DOI: 10.3390/agriculture11010050.
- Wedepohl, H K (1995). The composition of the continental crust. *Geochim. Cosmochim. Acta*, 59(7): 1217-1232. DOI: 10.1016/0016-7037(95)00038-2.
- Wilding, L P (1985). Soil spatial variability: Its documentation, accommodation and implications to soil surveys. *Soil spatial variability, workshop (Las Vegas NV 1984-11-30)*. Pudoc, Wageningen, Netherlands. p. 166-194.
- Wong, K W; Yap, C K; Nulit, R; Hamzah, M S; Chen, S K; Cheng, W H; Karami, A and Al-Shami S A (2017). Effects of anthropogenic activities on the heavy metal levels in the clams and sediments in a tropical river. *Environ. Sci. Pollut. Res.*, 24:116-134. DOI: 10.1007/s11356-016-7951-z.
- Xu, L; Zhao, Y; Wang, M and Jiang, Y (2021). Case study from Xuzhou area, Jiangsu, China. *Env. Pollut. Bioavail.*, 33(1): 326-333. DOI:10.1080/26395940.2021.1987842.
- Yang, Q; Li, Z; Lu, X; Duan, Q; Huang, L and Bi, J (2018). A review of soil heavy metal pollution from industrial and agricultural regions in China: Pollution and risk assessment. *Sci. Total Environ.*, 642(1): 690-700. DOI: 10.1016/j.scitotenv.2018.06.068.
- Zarcinas, B A; Ishak, C F; McLaughlin, M J and Cozens, G (2004). Heavy metals in soils and crops in Southeast Asia. 1. Peninsular Malaysia. *Environ. Geochem. Health*, 26(3): 343-357. DOI: 10.1007/s10653-005-4669-0.
- Zhang, B and Yang, Y (2017). Spatiotemporal modeling and prediction of soil heavy metals based on spatiotemporal cokriging. *Sci. Rep.*, 7: 16750. DOI: 10.1038/s41598-017-17018-5.

Zhang, H; Liu, W; Zhang, H; Fan, L and Ma, S (2019).
Spatial distribution of soil organic matter in a coal

mining subsidence area. *Acta Agric. Scand.*, 70(2): 117-
127, DOI: 10.1080/09064710.2019.1676916.

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