

## HEAVY METALS IN SOIL FROM BAUXITE MINING AREA OF KUANTAN, MALAYSIA

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### ABSTRACT

The community in the surrounding was reported serious air pollution from bauxite dust and residue that were released by the processing plants and during transportation around Bukit Goh, Gebeng and Kuantan Port, Malaysia. Bauxite contains trace quantities of mercury, hydrated aluminium oxides, hydrated aluminosilicates, iron oxides, titanium oxide, silica and other heavy metals. To measure heavy metals content in deposited soil in the settlement near bauxite mining area of Kuantan. Soil dust was grabbed in two sites near the mining area from December 2015 to February 2016. Samples were analysed with X-ray fluorescence (XRF) (HD Rocksand101). Analysis has detected 36 elements in the soil samples (N = 40). The highest non-carcinogenic element detected was Fe (194,912.75 ± 30,229.31 ppm) followed by Si (160,170.50 ± 41,784.35 ppm), Ti (26,740.40 ± 3,485.32), Ca (4,672.35 ± 4,042.28 ppm), Mn (1,551.65 ± 954.83 ppm) and Ba (1,260.88 ± 1,523.29 ppm). Mo (597.95 ± 82.05 ppm), Zn (212.90 ± 97.76 ppm) and Hg (36.33 ± 59.47 ppm) also was highly detected. Six carcinogenic elements detected were Cr (480.30 ± 61.47 ppm), Ni (87.30 ± 52.88 ppm), Pb (108.06 ± 78.88 ppm), As (25.17 ± 37.48 ppm), Cd (4.61 ± 0.78 ppm) and Se (1.87 ± 0.28 ppm). Traces elements with prominent value were Pr (534.29 ± 37.77 ppm), V (524.35 ± 71.00 ppm), Ce (222.95 ± 70.70 ppm), Nd (202.94 ± 79.89 ppm), Hf (91.24 ± 18.78 ppm) and Y (31.16 ± 14.84 ppm). Bauxite mining activity has impacted the soil in the studied area and produced probable health risk effects from the exposure.

**Keywords:** Bauxite, heavy metals, mining, soil, XRF

### INTRODUCTION

Bauxite is reddish clay with a pisolitic structure, earthy luster and a low specific gravity [1]. It is an ore formed leached of with other soluble materials from severely weathered rocks in a wet tropical and sub-tropical climate [2]. In tropical regions, lateritic bauxite or silicate Bauxites ores are largely formed by the weathering process of silicate rocks. These ores contain the highest concentration of aluminium compared to karst or carbonate bauxites [1].

The continuing demand for mineral supply nationally and globally has spurred the mining industry in Malaysia. Bauxite mining is among the metallic mineral sector in Malaysia includes iron ore, manganese, gold, tin, and other byproducts of tin and gold [3]. The 18,000 ha area in Kuantan (including Bukit Goh) is heavily mined for bauxite [4]. The Kuantan area is occupied with basalt composed of 12–13% Al<sub>2</sub>O<sub>3</sub>, 3–6% Fe<sub>2</sub>O<sub>3</sub>, 7–8% FeO, 1–2% TiO<sub>2</sub>, 0.02 % Cr<sub>2</sub>O<sub>3</sub>, and 0.01 % NiO [5, 6]. The soil of Kuantan Series contains gibbsite (Al(OH)<sub>3</sub>) mixed of goethite (FeOOH), hematite (Fe<sub>2</sub>O<sub>3</sub>), and kaolin [7, 8]. The presence of mineral hematite produces the red colour of the bauxite in Kuantan.

Bauxite is usually strip mined as it is typically found 1 or 2 m below the soil layer. Mining activities produced detrimental impacts to the environment, mainly the water, air, soil and human health [9]. The impact varies depending on whether it is an active or abandoned site, type of method being used and the meteorological and geological conditions [10]. Kusin et al., [11] highlights the bauxite mine-impact on sediments in Kuantan Port and Bukit Goh, where metals such as Al, Fe, Mn, As, and Pb were higher than the recommended guidelines values. The concentration of Cd (728.33 to 1811.66 mg/kg<sup>-1</sup>), Pb (323.66 to 802.66 mg/kg<sup>-1</sup>) and Zn (510.66 to 1097.33mg/kg<sup>-1</sup>) also determined above the background level in soils of the magnesite and bauxite mining waste dumps in Tamil Nadu, India [12]. Owing to the recent environmental degradation from the mining activities and the potential of its impact to public health, it is important that the occurrence of the pollution be further investigated. Therefore, this study was undertaken to evaluate the level of heavy metals in the soil samples from the bauxite mining area of Kuantan Port and Bukit Goh in Kuantan, Pahang. Evaluations were made by analysing the total content of heavy metals. This study provides baseline information on the level of contaminants in the soil as the effect of mining activity.

## METHODOLOGY

### Description of study area

Soil samples were collected at 40 sites in the vicinity of Bukit Goh (BG) (N = 20) and Kuantan Port (KP) (N = 20), including the stockpile areas that most affected by bauxite mining activities. Sampling was done from December 2015 to February 2016 during the temporary cessation of the bauxite mining activity (Figure. 1). Kuantan is a district in eastern Peninsular Malaysia, located at latitude  $3^{\circ} 45' 0''$  N, and longitude  $102^{\circ} 30' 0''$  E with an area of  $2,960 \text{ km}^2$ . Kuantan grow as a hub for trade, commerce, transportation, and tourism for the East Coast of Peninsular Malaysia due to its strategic location. The bauxite mining operation in Kuantan started in 2014 is progressively occurring in the vicinity of Bukit Goh. The ore deposits were transported to a temporary storage area within Kuantan Port prior to being exported to China for mineral processing [11].

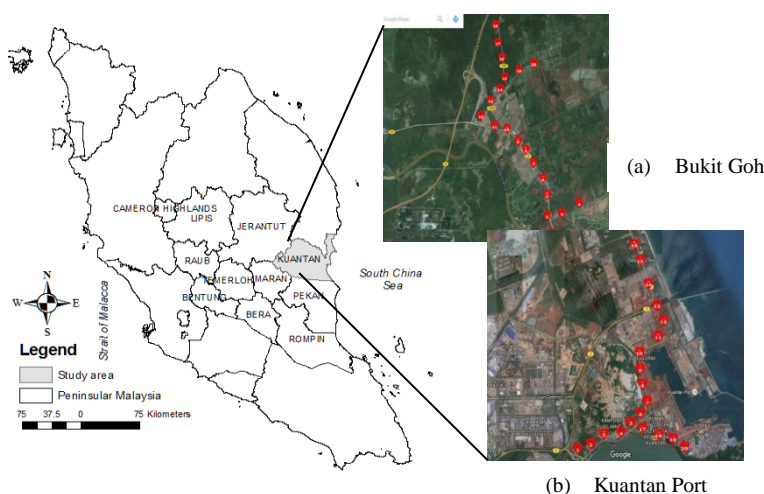


Figure 1. The sampling station of soil samples in (a) Bukit Goh and (b) Kuantan Port

### Samples preparation and analysis

Soil samples were air-dried at room temperature ( $28^{\circ}\text{C}$ ) and gently ground using a ceramic pestle and mortar and then passed through a number 50 sieve ( $<275 \mu\text{m}$  particle size) [13]. Samples were transfer to fill the sample cell and covert with an X-ray film. Samples were analyzed using high definition X-ray fluorescence (HDXRF®) HD Rocksand XOS's (Model 800701-01) for metal elements. The X-ray analysis detects emission from virtually all elements, depending on the wavelength and intensity of incident x-rays. XRF is a powerful quantitative and qualitative analytical tool for elemental analysis and ideally suited to measure film thickness and composition, elemental concentration by weight of solids and solutions, and identification of specific and trace elements in complex sample matrices. XRF analysis is used extensively in many industries including semiconductors, telecommunications and microelectronics. The method is fast, accurate, non-destructive, and requires only minimal sample preparation. The XRF machine was calibrated for each monochromatic beam and the detector. Calibration was done using multiple soil calibration standards consisting of background soil level, low contamination levels and high contamination level for selected elements to determine the initial sensitivity factor of each monochromatic beam. To ensure the quality of the results, a quality control sample (reference material representative of samples analyzed, NIST) is used for establishing and monitoring the stability and precision of an analytical measurement system. The percentage of recoveries for the metals studied ranged between 80 and 120%. A duplicate analysis of a sample was performed periodically to ensure precision of the sample analysis [14]. Data were analysed with SPSS.

## RESULTS

### Elements detected in samples

In total, 36 elements detected in the samples, itemized into carcinogenic, non-carcinogenic and traces elements (Table 1). The highest carcinogenic elements detected in most of the soil samples in both sites were Cr ( $480.30 \pm 61.47 \text{ ppm}$ ) followed by Pb ( $108.06 \pm 78.88 \text{ ppm}$ ), Ni ( $87.30 \pm 52.88 \text{ ppm}$ ), As ( $25.17 \pm 37.48 \text{ ppm}$ ), Cd ( $4.61 \pm 0.78 \text{ ppm}$ ) and Se ( $1.87 \pm 0.28 \text{ ppm}$ ). Among the highest non-carcinogenic element detected were Fe ( $194,912.75 \pm 30,229.31 \text{ ppm}$ ) followed by Ca ( $4,672.35 \pm 4,042.28 \text{ ppm}$ ), Mn ( $1,551.65 \pm 954.83 \text{ ppm}$ ) and Ba ( $1,260.88 \pm 1,523.29 \text{ ppm}$ ). Mo ( $597.95 \pm 82.05 \text{ ppm}$ ), Zn ( $212.90 \pm 97.76 \text{ ppm}$ ) and Hg ( $36.33 \pm 59.47 \text{ ppm}$ ) also was high in the soil samples. Traces elements that was highly detected in the samples

were Si ( $160,170.50 \pm 41,784.35$  ppm), Ti ( $26,740.40 \pm 3,485.32$ ), Pr ( $534.29 \pm 37.77$  ppm), V ( $524.35 \pm 71.00$  ppm), Ce ( $222.95 \pm 70.70$  ppm) and Nd ( $202.94 \pm 79.89$  ppm). Most of the elements were prominent in Kuantan Port compared to Bukit Goh.

## **DISCUSSION**

Of the environmental impact in mining area the major concern are related to the ore and waste mineralogy, the waste management during the mine operation and after decommissioning and the climatic conditions of the site (Razo, 2004). This study the first to report heavy metals contamination in soil after Kusin et al., [11] who measure the environmental impact of bauxite mining activity in Kuantan to sediments and water. The site studied is particularly interesting because of the recent public outcry over apparent environmental impacts associated with the bauxite mining activities, the lack of pollution control in recent mining operations and the influence of tropical climate conditions which the mine wastes are exposed to. Bauxite is not a mineral but it is a rock formed from a laterite soil that has been severely leached of silica and other soluble materials in a wet tropical or subtropical climate. There is no specific composition for bauxite rather it is a mixture of hydrous aluminum oxides, aluminum hydroxides, clay minerals, and insoluble materials such as quartz, hematite, magnetite, siderite, and goethite.

This study described the levels of heavy metals in soil of bauxite mining area in Kuantan, a month after the temporary cessation of the bauxite mining activities were announced by the authority. There are 36 elements detected in the soil sample, classified as elements with carcinogenic effects, non-carcinogenic and traces. Among the most prominent element detected in non-carcinogenic group were Fe, Mn, Ca, Ba, Mo, Zn and Hg. Carcinogenic elements Cr, Pb, Ni, As, Cd and Se were the highest detected. Most of these elements were prominent in Kuantan Port, where the mine wastes are stored. On the other hand, traces elements of Si, Ti V, Ce, Pr and Nd were also highly detected in this study.

These elements are possibly derived from the dust and aerosol emission of the mining operations and the mine waste in the stockpile area of Kuantan Port [11, 15]. The transport of metal dust from the mine wastes, along the prevailing wind direction cause particle grain deposited and segregated. This segregation may occur during wind events resulting in re-suspension of coarse particles from the mine wastes. Once the coarse particles are deposited within meters of the mine waste deposits (Kuantan Port), the fine fraction of this metal dust may be transported to long distances (to Bukit Goh) by winds. This possibly causes direct effect on the population around mining area [15, 16]. Mine wastes are the source of heavy metal particles (e.g. pyrite, chalcopyrite, sphalerite, galena and arsenopyrite) giving rise to a typical suite of hazardous trace elements (Zn, Cu, Sb, Bi, Pb, As, Sn, Mo, Fe, Ni, Cd) with potentially adverse environmental effects to surrounding soils and plants [16]. The particles of mining and metallurgical operations are often dispersed by wind and or water after their disposal could be a significant source of metals release as residues in this study [17]. Climatic effects such as wind and heavy rainfalls have greater impact in the dispersion of metals in soils are typically sparsely vegetated. Wind erosion can be a major cause of the loss and dispersion of material from tailings impoundments into their surroundings and can affect tailings dams in all kinds of climates [18].

Although high metals concentration is an indication of potential anthropogenic influence, some natural sources can also cause the observed enrichment (Atgin et al. 2000). This mineral and local emission factor comprises a mixture of species caused by both natural origin (soil particle deposited) and anthropogenic origin (traffic, domestic emissions principally) [16]. Suspended mine waste dust contributes notably to the total concentrations of trace metals into the atmosphere (32%), although the mass concentrations of potentially toxic trace elements (Bi, As, Cu, Pb, Cd, Zn and Sb) are low. Other principal source contributions are crustal (33%) and regional ore are a mixture of industrial or fuel-oil combustion and secondary inorganic compounds (25%). Wind directions also favouring the transport of suspended particles towards the sampling station with the consequent impact on soil quality [15]. Agriculture activity and palm oil plantation in the study area also may contribute to the enrichment of these metals in soil from the use of fertilisers [11, 19]. The soil pH also plays an important role in metal mobility in the study area [20]. Metals such as Pb has low mobility in neutral or alkaline soils due to the formation of insoluble salts, while As, Cu and Zn, have greater mobility, due to the relative solubility of the complexes formed in neutral or alkaline soils.

According to Fernandez-Caliani et al.,[20], the anthropogenic elements Bi, As, Cu, Pb, Cd, Zn and Sb are derived from pyrite-bearing materials and mine soils. High Mn and Nd shows that the source of these elements are related to bauxite mine, as silicate mineral is one of the mixture in bauxite. Cr, V, Ti and Fe elements are derived from basalt. Bauxite in Kuantan area is formed from basalt (12–13% Al<sub>2</sub>O<sub>3</sub>, 3–6% Fe<sub>2</sub>O<sub>3</sub>, 7–8% FeO, 1–2% TiO<sub>2</sub>, 0.02 % Cr<sub>2</sub>O<sub>3</sub>, and 0.01 % NiO) [5, 6]. In addition, these metals also may derive from dust and aerosol emissions commonly particulates from mining operations and usually most concentrated in the finer particle size fraction, which travels greater distance in the environment. These elements also associated with significantly elevated levels of Hg, Pb, and As [21-24]. Besides, Ti, V and Fe are the elements of the silicate particles minerals resulting from the process of extraction, transport and re-suspension of soil particles in the mining area. These elements derived from parent rocks in the upper continental crust [15]. Iron (Fe) can be attributed to the iron oxides minerals soil of the Kuantan series that contains gibbsite (Al(OH)<sub>3</sub>) mixed of goethite (FeOOH), hematite (Fe<sub>2</sub>O<sub>3</sub>), and kaolin [7, 8]. The presence of mineral hematite produces the red color of the bauxite in Kuantan, existence of Fe. V is also can be attributed to particles emitted from some combustion plants, with long-range transport and high residence time in the atmosphere.

Elements such as K, Zr, Zn and Ca can be interpreted as crustal minerals resulting from the processes of extraction, transport and re-suspension of soil particles in the mining area [15]. Zn is commonly resulted from anthropogenic sources such as non-ferric metal industry and agricultural practice. Contemporarily observed soil contamination with Zn has already brought Zn to an extremely high in soils around mining areas as follows; Netherlands, 1020 mg/kg; Austria, 8900 mg/kg; and Greece, 10547 mg/kg [25]. Soils surrounding a Zn smelter in Poland contain Zn within the range of 202–4832 mg/kg, at an average of 1062 mg/kg [26]. Elevated Zn content of natural source (from bauxite parent material), at the range of 126–683 mg/kg, is reported for Oxisols in Jamaica [27].

**Table 1.** Concentration of elements that can cause carcinogenic health risk (ppm) in soil samples (N = 40)

Elements	Bukit Goh <sup>(n=20)</sup>				Kuantan Port <sup>(n=20)</sup>			
	Detected	Mean	Std. Dev	Range	Detected	Mean	Std. Dev	Range
<b>Elements with carcinogenic health risk</b>								
Cr	20	480.30	61.47	357 - 634	20	465.35	144.37	176 - 703
Ni	20	87.30	52.88	50 - 238	20	65.94	18.29	30 - 115
Pb	19	44.76	22.63	9 - 114	20	108.06	78.88	28 - 336
As	13	10.52	6.84	4 - 30	18	25.17	37.49	2 - 143
Cd	3	3.63	0.49	3 - 4	12	4.61	0.78	4 - 6
Se	13	1.87	0.28	1 - 2	5	2.04	0.21	2 - 2
<b>Elements with non-carcinogenic health risk</b>								
Fe	20	164,771.65	23,987.08	120,072 - 225,986	20	194,912.75	30,229.31	103,813 - 246,323
Mn	20	641.10	129.77	431 - 1022	20	1,551.65	954.83	524 - 4582
Mo	20	597.95	82.05	445 - 757	20	546.40	79.08	370 - 650
Sr	20	34.83	10.77	13.50 - 53.70	20	43.00	26.24	25.50 - 141
Sn	11	33.72	17.71	14.60 - 69.60	14	66.26	58.02	15.80 - 205
Tl	15	3.60	0.56	2.40 - 4.50	17	4.76	1.17	3.20 - 7.30
Ca	20	4,672.35	4,042.28	367 - 16,949	20	2,564.55	1,953.92	409 - 7,347
Ba	13	695.08	164.68	408 - 886	16	1,260.88	1,523.29	510 - 6903
Cu	20	90.86	25.00	66.5 - 154	20	100.09	32.79	57.70 - 206
Hg	3	1.97	0.21	1.8 - 2.2	3	2.63	0.40	2.20 - 3.00
Zn	20	171.80	63.17	105 - 354	20	212.90	97.76	93.90 - 503
Ag	3	5.50	0.52	5.20 - 6.10	4	5.58	0.35	5.20 - 6.00
<b>Elements considered as tracers</b>								
Si	6	160,170.50	41,784.34	86,031 - 197,889	12	159,239.25	49,229.02	86,127 - 285,259
Ti	20	26,740.40	3,485.32	20,976 - 32,780	20	21,827.75	8,062.05	7,670 - 36,259
K	17	3,841.82	2,325.47	158 - 7,688	20	3,528.95	2,746.68	445 - 12,076
V	20	524.35	71.00	372 - 640	20	442.30	153.79	168 - 741
Pr	20	457.70	82.69	321 - 619	17	534.29	37.77	468 - 605
Zr	20	412.35	109.98	280 - 637	20	392.30	123.03	251 - 721
Ce	20	165.60	38.78	101 - 256	20	222.95	70.70	128 - 364
Nd	15	127.03	34.13	80.80 - 209	16	202.94	79.88	101 - 411
Hf	20	80.32	13.89.917	66.60 - 119	20	91.24	18.78	33.90 - 130
Ga	20	44.12	6.98	35.50 - 60.60	20	46.22	9.42	29.30 - 66.90
Rb	16	38.18	20.24	7.50 - 72.30	16	24.09	23.41	4.10 - 98.80
W	20	31.30	6.80	14.10 - 38.10	19	39.75	12.26	9.40 - 64.40
Y	20	22.91	11.87	9.60 - 45.60	20	31.16	14.84	9.20 - 52.50
Br	19	8.52	2.71	4.20 - 15.40	16	8.66	1.93	5.40 - 14.10
Rh	5	6.12	1.29	4.80 - 7.80	7	5.21	0.891	4.10 - 6.60
Bi	9	5.19	1.54	3.70 - 8.80	15	12.15	12.35	5.20 - 55.30
Pd	12	5.19	0.93	4 - 6	15	5.69	0.86	4.20 - 6.90
Ge	15	2.01	0.58	1.20 - 3.50	15	1.95	0.640	1.10 - 3.20

Note: BDL – below detection limit

Ca is commonly found in sedimentary rocks in the minerals calcite, dolomite ( $\text{CaMg}(\text{CO}_3)_2$ ), and gypsum ( $\text{CaSO}_4 \cdot \text{H}_2\text{O}$ ). It also occurs in igneous and metamorphic rocks chiefly in the silicate minerals: plagioclases, amphiboles, pyroxenes, and garnets. Calcite is the most widespread and relatively mobile form of Ca carbonates present in soils. It is greatly dispersed and has a major influence on the pH of soils and therefore on trace element behaviour in this study. Carbonates of Ca and Mg are inherited mainly from limestones, but may also form in soils, under specific conditions (neof ormation processes) [25].

Mo, Ce and Rb are mainly regarded as crustal minerals from the mining process. The most common Mo mineral is molybdenite ( $\text{MoS}_2$ ), which occurs in several geological formations. Less frequent are molybdenite ( $\text{MoO}_3$ ), wulfenite ( $\text{PbMoO}_4$ ) and Powellite ( $\text{CaMoO}_4$ ). Several other Mo minerals, mainly oxides are known to be associated with various sediments and often are associated with Fe and Ti minerals. Mo is a primary ore deposit and a by-products of copper mines [25]. Cerium (Ce) is a lanthanide (LA) a rare earth elements (REE) which occur naturally in the Earth's crust. This element is being incorporated in relatively common minerals such as monazite, bastnasite, cheralite and xenotime which is often associated with phosphatic rocks resulted in elevated Ce in phosphorus fertilisers. Rubidium (Rb) is considered to be a dispersed element from the mining process [28] and does not form minerals of its own. The main source of Strontium (Sr) pollution is associated with coal combustions and sulfur mining [25].

## **CONCLUSION**

This study has determined 36 elements in the soil sample. Most of the elements were highly detected in Kuantan Port, a bauxite stockpile area. Elements detected were possibly derived from the mine waste or residues as well as dust and aerosol emission from the mining operations. It also possibly related to crustal mineral and mine residues resulting from the extraction, transportation and deposited of soil particles in the mining area.

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