PRELIMINARY STUDY OF HEAVY METAL (Zn, Pb, Cr, As, Cu, Cd) CONTAMINATIONS ON DIFFERENT SOIL LEVEL FROM POST-MINING BAUXITE PRODUCTION FOR AQUACULTURE

Risandi Dwirama Putra[1*], *Tri* Apriadi[2], *Ani* Suryanti[3], *Henky* Irawan[4], *Tengku* Said Raja'I[4], *Try* Yulianto[4], *Wiwin* Kusuma Atmadja[4] and *Chandra* Joei Koenawan[1]

[1] Marine Science Departement, Faculty of Marine Science and Fisheries, Raja Ali Haji Maritime University, Jl. Politeknik, Senggarang 29111, Tanjungpinang, Indonesia
[2] Aquatic Resources Management Departement, Faculty of Marine Science and Fisheries, Raja Ali Haji Maritime University, Jl. Politeknik, Senggarang 29111, Tanjungpinang, Indonesia
[3] Center/Division/Laboratory, Institution, City with Postal Code, Country
[3] Sosio-Economic Departement, Faculty of Marine Science and Fisheries, Raja Ali Haji Maritime University, Jl. Politeknik, Senggarang 29111, Tanjungpinang, Indonesia
[4] Aquaculture Departement, Faculty of Marine Science and Fisheries, Raja Ali Haji Maritime University, Jl. Politeknik, Senggarang 29111, Tanjungpinang, Indonesia

Abstract Bauxite production and exploration give negative impact environmental modification and increasing concern pollution of heavy metals. One of an alternative to reduce the negative impact of bauxite post-mining production is by re-utilizing the abandoned bauxite land and necessary exploratory study to obtain the current environmental conditions primarily related to heavy metals in bauxite mining area. Soil and water quality samples from 5 (five) sites based on post-mining activities were used to determine concentration heavy metals of As, Cd, Pb, Zn, Cr, Cu from different soil level. They were also collected for grain size analysis including those of sand and mud percentages along with the mean, and sorting analysis. The mean grain size post-mining bauxite ranged between very fine sand to medium sand and had heterogeneous texture. The coarse grain size percentage increased towards the bottom of the soil. Total heavy metal contents for post-mining bauxite soils are 0,081 ppm, 0,245 to 0,471 ppm and 0,007 ppm for As, Pb and Cd respectively. Heavy metals for Cd, uniformly at every depth from soil level. On the other hand, Pb showed significance pattern, it was indicated every depth from soil level, the concentration of Pb was different.

[*] Corresponding author, e-mail: yanti.ajb@umrah.ac.id

1. INTRODUCTION

Many pollutants are accumulated in the sediments and have harmful effects on the aquatic environments [1]. The sediments of the aquatic environment act as a major reservoir of heavy metals and source of contaminants [2]. Heavy metals are natural constituents of sediment and their concentration varies depending on parental materials [3]. Heavy metals can be present in the soil as a product of the weathering of the natural rocks, or because they come as part of pollution loads generated by human activities [3].

In many cases of miner exploration, the action of the water over tailings and other miner wastes causes a phenomenon known as acid mine drainage (AMD) [4]. The activities developed by bauxite mining are, from the very start of operations, capable of degrading the surrounding environment and causing major environmental modification [4]. Several areas which are former bauxite mining are damaged and give negative impact environmental modification. The process of dredging bauxite in some mining sites resulting in the emergence of puddles as well as leaving a barren and unproductive land.
Some research related to bauxite mining has been studied by researcher and almost researcher generally investigated on the impact of bauxite tailing/red mud. Inappropriate storage of red mud, a by-product of alumina production, is of increasing concern, as illustrated by a major pollution incident [5].

The average metal concentration from the largest to the smallest was Fe, Ni, Mn, Zn, Pb, Cu, Cd, and Cr. The value from results of metal concentration in bauxite sample was still below from the specified standard quality threshold. Bauxite has a variety of important minerals, including iron, silica, titanium, calcium, and magnesium. Further complementary minerals contained in bauxite were Na, K, P, Cr, V, Ga, Zr, Zn, Pb, Cu, Ni, Mn and Co [6] It is further explained that the main chemical content of red mud from bauxite tailing consists of Fe2O3 (48.50%), SiO2 (11.53%) and Al2O3 (14.14%) and additives of TiO2 (5.42%) and V2O5 (up to 0.116%). Zr element has the largest proportion element that contained in bauxite samples ranging from 460-1790 ppm and abundant element found in next is V element with a range of 210-416 ppm then Cr with a range of 108-326 ppm. Ni, Cu, Zn, Rb, Sr, Y, Nb, Ce, La, and Pb are found in small amounts (<100 ppm) [7]. Inappropriate storage of red mud, a by-product of alumina production, is of increasing concern, as illustrated by a major pollution incident[6]. Red mud can also contain problematic concentrations of potentially toxic metals and metalloids, including As, Cr, Ni, Pb, Mo and V (Brunori et al., 2005; Grafe et al., 2011; Klebercz et al., 2012) and contains several potentially toxic elements, including arsenic [8].

The most common characteristic of exploration and production mining on bauxite give the impact destruction of soil which allows the erosion and causing the topsoil to become thinner or even lost and bauxite extraction clearly disturbs the upper regolith and eliminates any indurated layer present [9]. The former or primary soil of bauxite mines usually dense and difficult to process because of the structure, texture, porosity, and bulk density of primary soil that affects root system development and disrupts plant growth [10].

One of an alternative to reduce the negative impact of bauxite mining production is by re-utilizing the abandoned bauxite land. Excavations of bauxite mining will generally be filled by water (generally rainwater) that formed a puddle of water. Puddles on the former bauxite excavations are thought to have the potential for fish farming. Potential land for bauxite excavation for fish cultivation has not been studied before Therefore, it is necessary exploratory study to obtain the current environmental conditions primarily related to heavy metals in bauxite mining area and concentration of macro and micro minerals concentrate that function as nutrients in the process of fish farming. Based on information on the concentration of minerals and related metals contained in the bauxite land will be known potential development of future utilization of bauxite land.

2 MATERIALS AND METHODS

2.1 Sampling and sediment collection

Each sampling location was determined using a GPS and located from 5 (five) sites based on post-mining activities (Table 1) were used to determine concentration heavy metals of As, Cd, Pb, Zn, Cu, Cr from different soil level (0-30 cm), (30-60 cm) and (60-100 cm). They were also collected for grain size analysis including those of sand and mud percentages along with the mean, and sorting analysis [11]. The concentration of total heavy metals was measured using Flame-Atomic Absorption Spectrophotometer using AA-6200 Shimadzu, 10 mA currents by adjusting the specified wavelengths; acetylene air gas 21/min; O_2 15 L /min, temperature 2000°Cc.

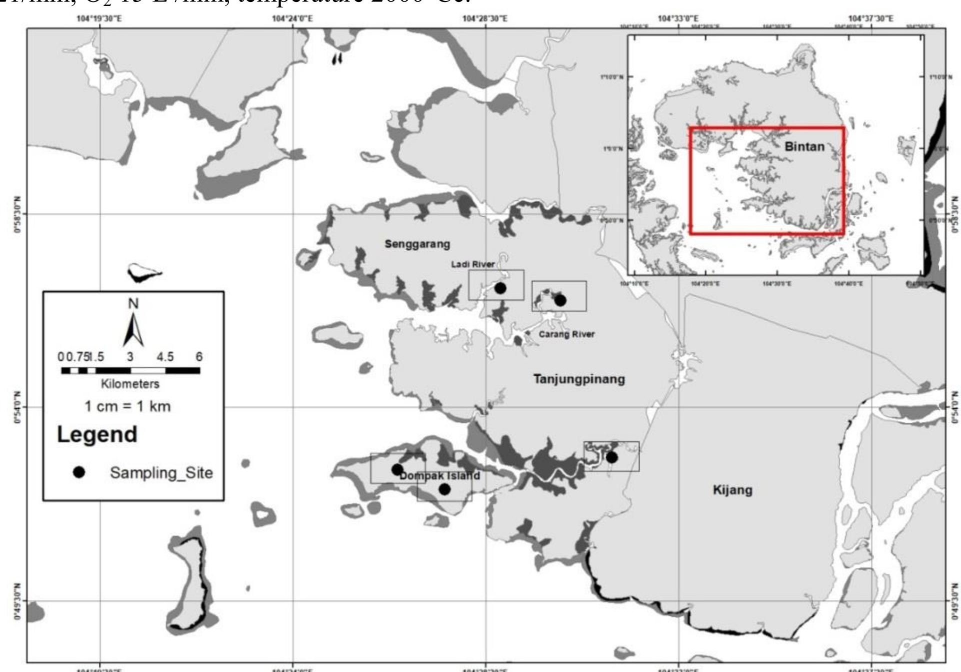


Fig 1. Map of Study area

Table 1. Geographic locations of sampling site and data of collecting cores samples

Sampling Site	Location		Soil depth (cm)	Water Depth (cm)
	Latitude	Longitude		
Sei. Ladi	0.94636	104.4807	100	37
Sei. Carang	0.94169	104.5041	100	28
Simpang Dompak	0.87582	104.4406	100	10
Dompak	0.86830	104.4591	100	21
Wacopek	0.88071	104.5240	100	8

2.2 Sediment core and water quality parameter instrument

15 sediment cores for the laboratory experiment were collected within an area of post-mining bauxite production. The cores had an internal diameter 65 mm and consisted of 100 cm depth of sediment [5]. DO concentration. conductivity. pH and water temperature were measured in *situ* at the sample site and in the collected sediment cores using a SensoDirect 150 Lovibond® Portable Multi-Parameter Meter and Probes.

2.3 Dilution of Sediment for trace metals analysis

Chemical Acid (70% HNO_3). Hydrofluoric acid (HF) and deionized water were used for dilution. Blank samples and quality control standard for trace metals analysis [12].

2.4. Sample Preparation

Sediment samples were prepared according to an adapted USEPA 3050B method to determine the total heavy metals for Cr. Mn. Fe. Cu. Zn. and Pb in core sediments [35]. The method was altered by using approximately 0.20 g of the sample as described by [13]. Briefly. Samples were dried in an oven at 105^{o}C and ground into a powder using a mortar and pestle. Nitric acid (70% HNO3) and hydrofluoric acid (HF) were used for the total metal dissolution. Sediment samples were digested in heavy acid with 3:1 mixture of HNO3 and HF acid. Nine ml of HNO_3 and three ml of HF were added to the sediment samples inside a fume hood. Sediment samples plus HNO3 and HF acid were digested at 100^{o}C on a hot plate until dryness [12].

2.5 Enrichment Factor (EF)

The Enrichment Factor (EF) in metals and Geo accumulation Index (I_{geo}) are indicators used to assess the presence and intensity of anthropogenic contaminant deposition on surface soil [3]. The EF calculation seeks to reduce the metal variability associated with variations in mud/sand ratios and is a convenient tool for plotting geochemical trends across large geographic areas. Which may have substantial variations in the mud (i.e. clay-rich) to sand ratios [14]. For assess the possibility of anthropogenic influence in the study area. EF was calculated for the investigated three total heavy metals. The essential reason for using geochemical studies is to determine differences of metals from anthropogenic activities and those from natural sources [12]. The EF method normalizes the measured heavy metal content with respect to a sample reference metal using Fe based on [14]. Fe as an acceptable normalization element to be used in the calculation of the enrichment factor since they considered the Fe distribution was not related to other heavy metals. The EF is calculated according to the following equation:

$$EF = M_x \text{ x } Fe_y / M_b \text{ x } Fe_b \quad (1)$$

Where M_x and Fe_x are the sediment sample concentration of the heavy metal and Fe [14]. while M_b and Fe_b are their concentrations in a suitable background or baseline reference material [15]. Hence. for quality classification. EF results are applied to a qualitative scale of metal concentration based on [16] (*reference levels; <2 deficiency to minimal enrichment ; 2-5 moderated enrichment; 5-20 significant enrichment.; 20-40 very high enrichment ;> 40 extremely high enrichment*).

$$I_{geo} = \log_2 C_n / 1.5 B_n \quad (2)$$

Where Cn is the concentration of the element in the enriched samples. and the Bn is the background or pristine value of the element. Factor 1.5 is introduced to minimize the effect of possible variations in the background values, which may be attributed to lithologic variation in the sediments [1,14]

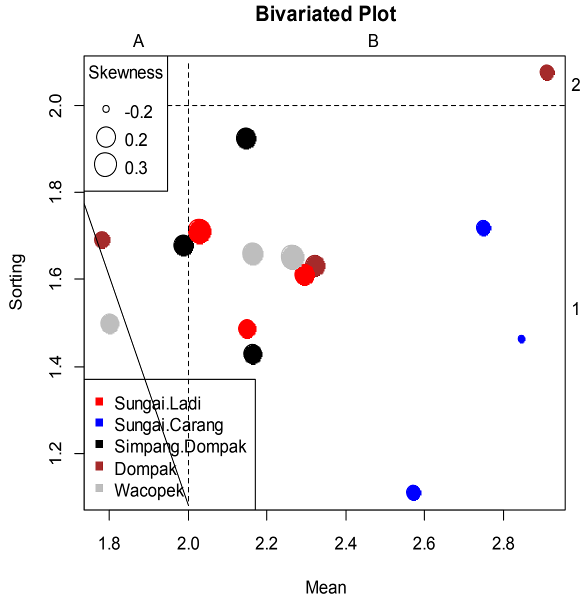
Hence. for quality classification. I_{geo} results are applied to a qualitative scale of enrichment intensity based on [17] (*reference levels; 0-1 uncontaminated; 1-2 unpolluted to moderately contaminated; 2-3 moderately contaminated; 3-4 moderately to strongly contaminated; 4-5 strongly contaminated;>5 very strongly contaminated*) and. according to this scale. samples with I_{geo} above 1 show signs of enrichment/contamination. [1,2,18]

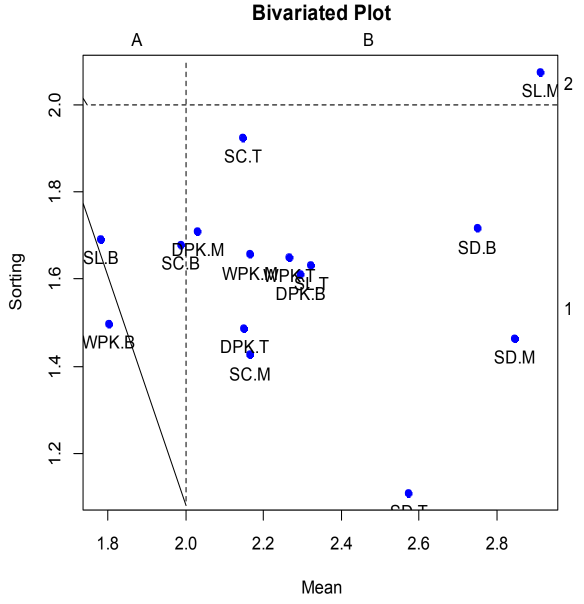
2.6 Statistical Analysis

The statistical analysis measurements using Bivariate-Plot sediment analysis to calculate mean grain size, degree of scatter of sorting, kurtosis and the degree of skewness as describe by [11, 19, 20, 21) using packages rysgran for R statistical analysis [22]. Sorting or standard deviation measures of post-mining bauxite sediment indicates the fluctuations in anthropogenic conditions of the tailing and excavating condition on bauxite mining. Graphical Anova to calculate variance based on post-mining bauxite sediment with One-way ANOVA analysis using packages granova for R statistical analysis [23], Effect of heavy metal contamination for aquaculture water quality measure using Principle Component Analysis (PCA) with multivariate data from pH, dissolved oxygen, temperature water column and all heavy metals tracer (Zn, Pb, Cr, As, Cu, Cd) based on confidence ellipse around the categories of site location [35]. Linear regression analysis from heavy metal tracer calculated using CCM (Coefficient Correlation Matrix) based on person method every sediment layer.

3 RESULTS AND DISCUSSION

3.1 Textural characteristics of sediment





Description

Mean	Verbal.Mean	Sorting	Verbal.Sorting
A	Medium sand	1	Poorly sorted
B	Fine sand	2	Very poorly sorted

Fig.2. Bivariate-Plot of sediment between Mean Size and Sorting Post-mine Bauxite Bintan Island.

The statically analysis reported cumulative probability curve provide skewness, kurtosis, sorting and mean size for each sediment sample (Fig. 2). The post-mining bauxite production in Bintan Island show textural facies of sediment dominated by sand (medium sand and fine Sand). Overall, most of every layer sediment enriched with fine sand and has relatively high sorting values categorized by poorly sorted. Some of sample sediment in bottom layer with medium sand (Sungai Ladi, Wacopek and Sei.Carang) and one sample categorizes in very poorly sorted (medium layer in Sei Ladi site). Kurtosis analysis show that all sample are better sorted at the tail than the central portions (approximately symmetrical), hence the mesokurtic curves. The sample obtained in surface layer show variation in the mean grain size between ϕ 2.14 to ϕ 2.57 and are categorized as fine sand. The sediment is poorly sorted, with standard deviation as high as 1.93. Bottom layer on bed material sediment on the hand has larger grain size, varying from ϕ 1.78 to ϕ 2.75 falling categorized medium sand to find sand. The mean size in bottom layer from post-mining bauxite sediment show consistent decreasing grain size toward sediment layer. Grain size distribution of post-bauxite sediment are generally associated with vertical sediment layer. In view of this, the small particles of sediment dominated in surface layer while the larger particles concentration in bottom layer.

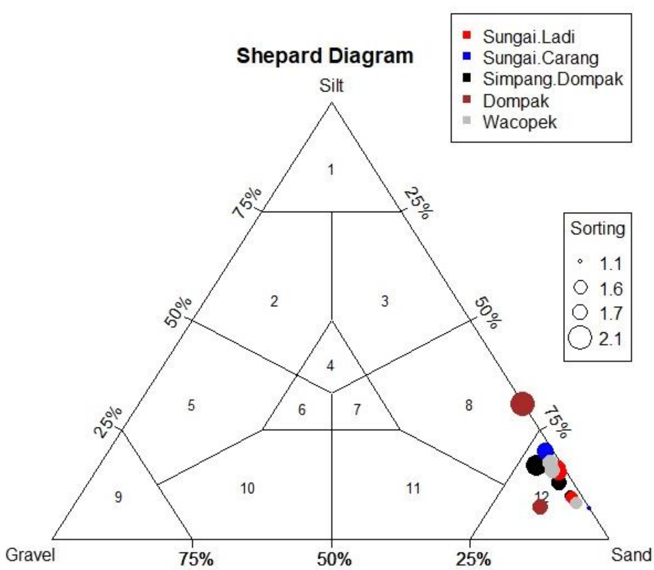


Fig 3. Shepard post-mining Bauxite diagram of Bintan Island.

The sum of these three basic component should be equal to 100% in Shepard triangle classification system. In study are (Fig 3), two kinds of sediment types were found, they were dominated categorized sand and only one site sampling categorized sandy silt which was in accordance with field survey result. The average percentage of sand was 89.92% and Silt was 14.97%.

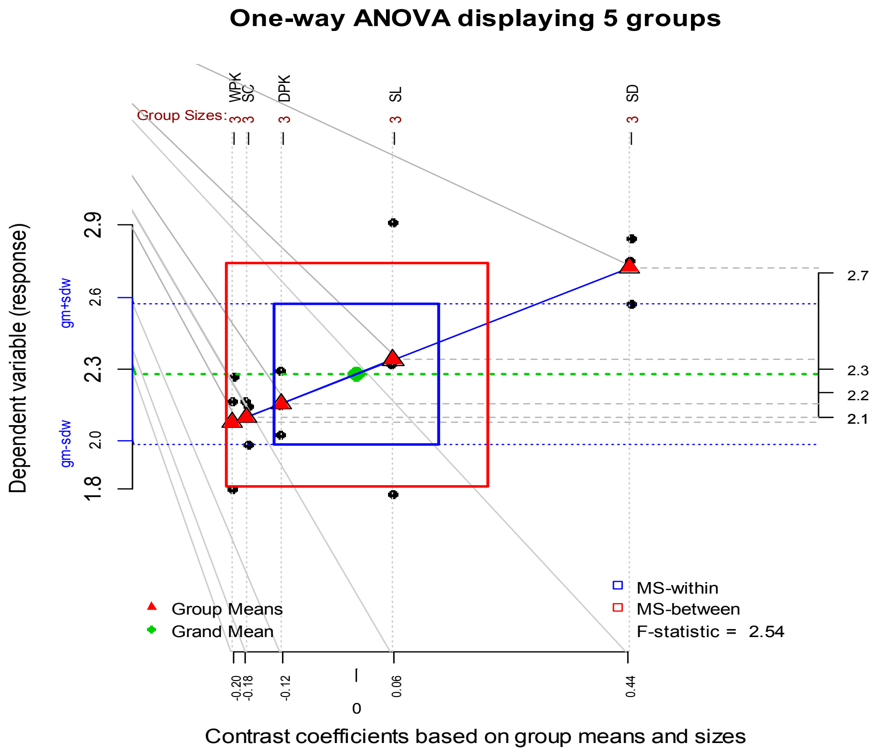
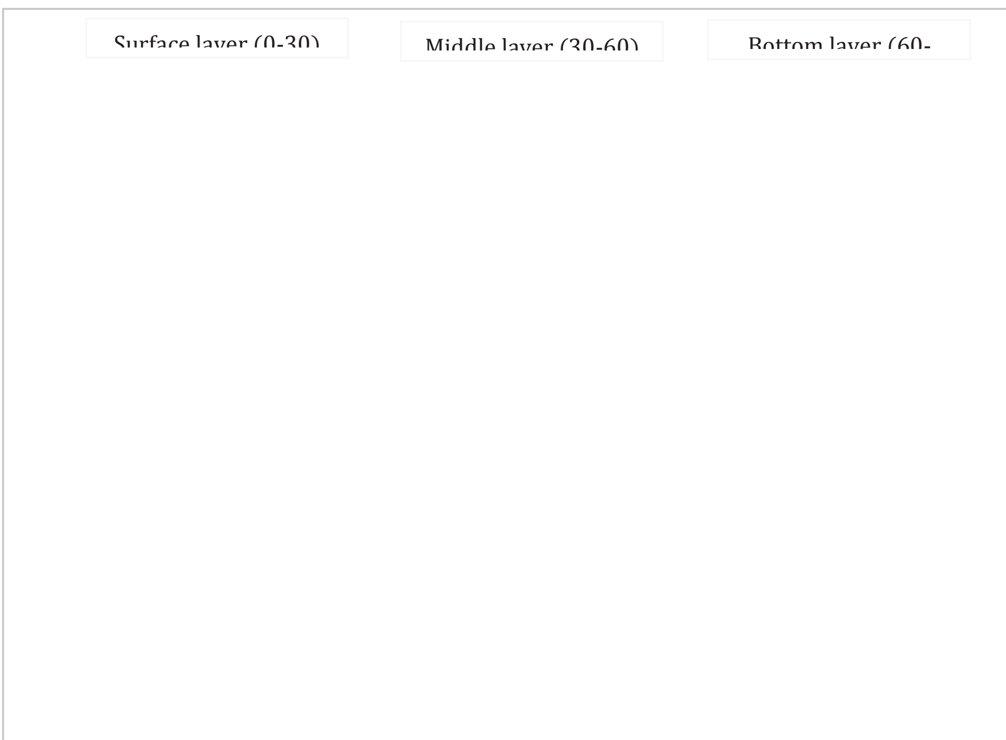


Fig 4. Graphical Anova post-mining Bauxite soil diagram of Bintan Island.

Comparison of 5 site sediment sampling in post-mining bauxite production, summary measure of mean ϕ and percent sediment resulted in same conclusion, there were sand category sediment. The estimated mean ϕ was equivalent across sediment layer for all sample (Fig 4). In addition, estimates of percent grain size ϕ non-significantly different among site and sediment layer (f-test 2.54 and p-value <0.05). The measurement variance non-differed over the range of half-ϕ intervals for all sediment site and layer. Summary on variance analysis show the highest grand-mean of sediment grain size from Simpang Dompak Site with mean ϕ value 2.7. In other hand, the lowest grand-mean of sediment grain size from Wacopek site with mean ϕ value 2.1. The highest and lowest contrast coefficient based on site group mean and size was 0.44 and 0.20 respectively.

3.2. Enrichment Factor (EF)

The result of Enrichment Factor of Heavy metals in Post-mining Bauxite from different soil level are represented in **Fig.5**. In the present study, Fe has been used as a conservative tracer to differentiate natural from the anthropogenic component. The results are indicated that the EF of Zn ranged from 6.93 to 42.51, which is significant to very high enrichment, respectively. The EF for Cd ranged between 6.15 and 6.26, which is significant enrichment. The EF of Pb varied from 32.55 to 63.25, which is very high enrichment to extremely high enrichment. The EF of Cu varied from 2.25 to 14.49, from 0.53 to 16.03, and from 107.61 to 206.29 for As. These result indicate that the EF for all of the studied metals is in moderated enrichment to significant enrichment except for Arsenic (As), which ranged extremely high enrichment. These heavy metals are classified in the sediment of post-mining bauxite by their source into a natural source (<1.5) and anthropogenic source (>1.5). Most of these metals come from the surrounding anthropogenic activities and exploration bauxite mining. The sequence of EF for heavy metals in the sediment of post-mining bauxite is in the following order: As > Pb > Zn > Cr > Cu > Cd. This indicates that arsenic was more abundant when compared with the others metals, where cadmium had the lowest appearance. An average enrichment factor of different soil level post-mining bauxite are relatively similar for surface level (0-30) cm, middle level (30-60) cm, and bottom level (60-100) cm. The obtained result of EF calculations showed Zn, Cd, Mn, Cu, and Cr to have EF values lowest in middle level of soil except for Pb which is lowest in surface level. Almost average enrichment factor from heavy metal had the highest value in bottom level.



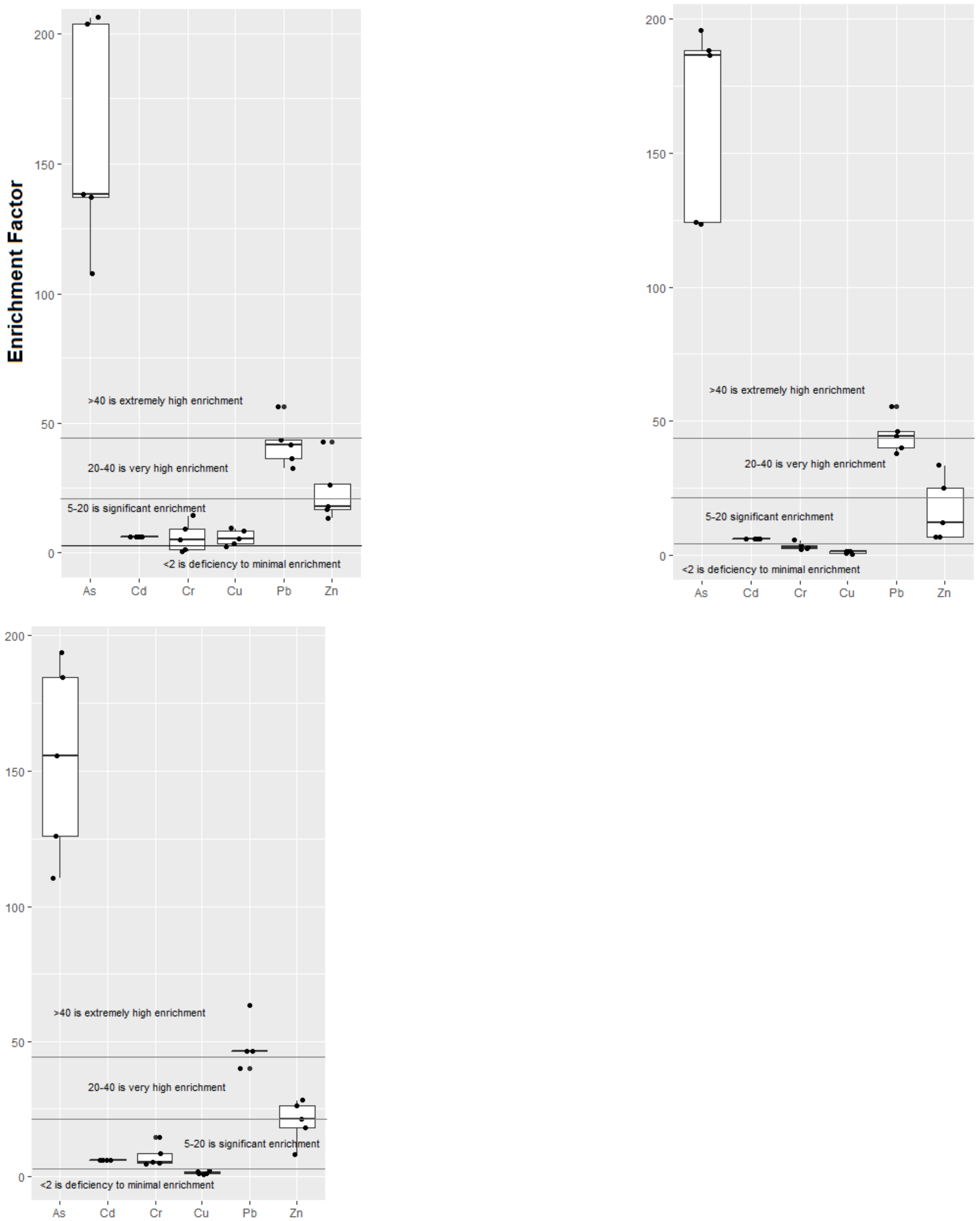


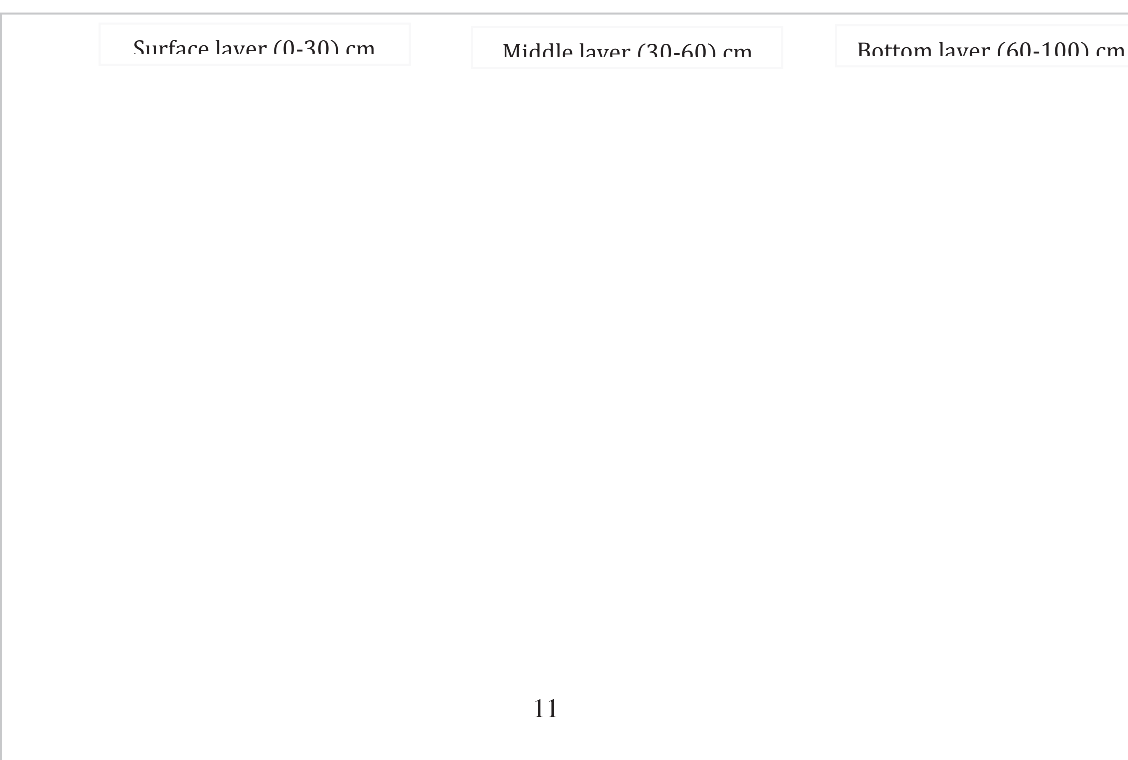
Fig 5. Enrichment Factor of heavy metal on post-mining bauxite from different soil level.

3.3 Geo-accumulation index (I_{geo})

The results of I_{geo} are as shown illustrated in Fig. 6. The positive values of heavy metal > 1.5 depending on the classification based on [17] indicated that the post-mining bauxite in every level of soil is polluted with this metal and used to quantitatively measure metal pollution in sediments based on a pollution intensity classification [16]. I_{geo} values showed very strongly contaminated pollution with arsenic (As) and lead (Pb) at every level of soil from post-mining bauxite production. For zinc (Zn), the values of I_{geo} showed moderate to strongly contaminated pollution degree. On the basis of above said classification, Cr and Cu in both showed uncontaminated to moderately pollution load.

The characteristic of sediment in bauxite mining is by having a red color [24,25,26] Presence of arsenic natural source in the sediment with red color provides lowest concentration of As, where black sediment produces mostly As. [27]. The result of enrichment factor of arsenic which ranged extremely high enrichment EF (107.61 to 206.29) was described as a mining exploration which contains huge amounts of wastes that may increase the amounts of Arsenic [2]. The high concentration of arsenic in the environment is suspected due to bauxite mining activities. High arsenic levels can also come from industrial activities such mining also contribute to arsenic in the environment [28]. The total content of Pb was indicative of enrichment at every level soil. Enrichment Pb indicating that the sediments were polluted [29] and allegedly originated consequently may cause significant ecological effect on the sediment surface (Desert et al., 2017). Enrichment lead (Pb) on sediment indicates the anthropogenic influence due to deposition and absorption by dredging on tailing process and disposal gas and oil on heavy equipment bauxite mining [30]. The enrichment of Zn appear to be present in the bottom level in amounts, which is explained by the high adsorption capacity of ground because Zn had correlation mainly in smaller grain size [31].

In summary, concentrations of Zn, Pb and As in sediment depended largely on the distance away from the mining area, or more specifically, the farther away from the mining area, the lower the concentrations of heavy metals [32] .



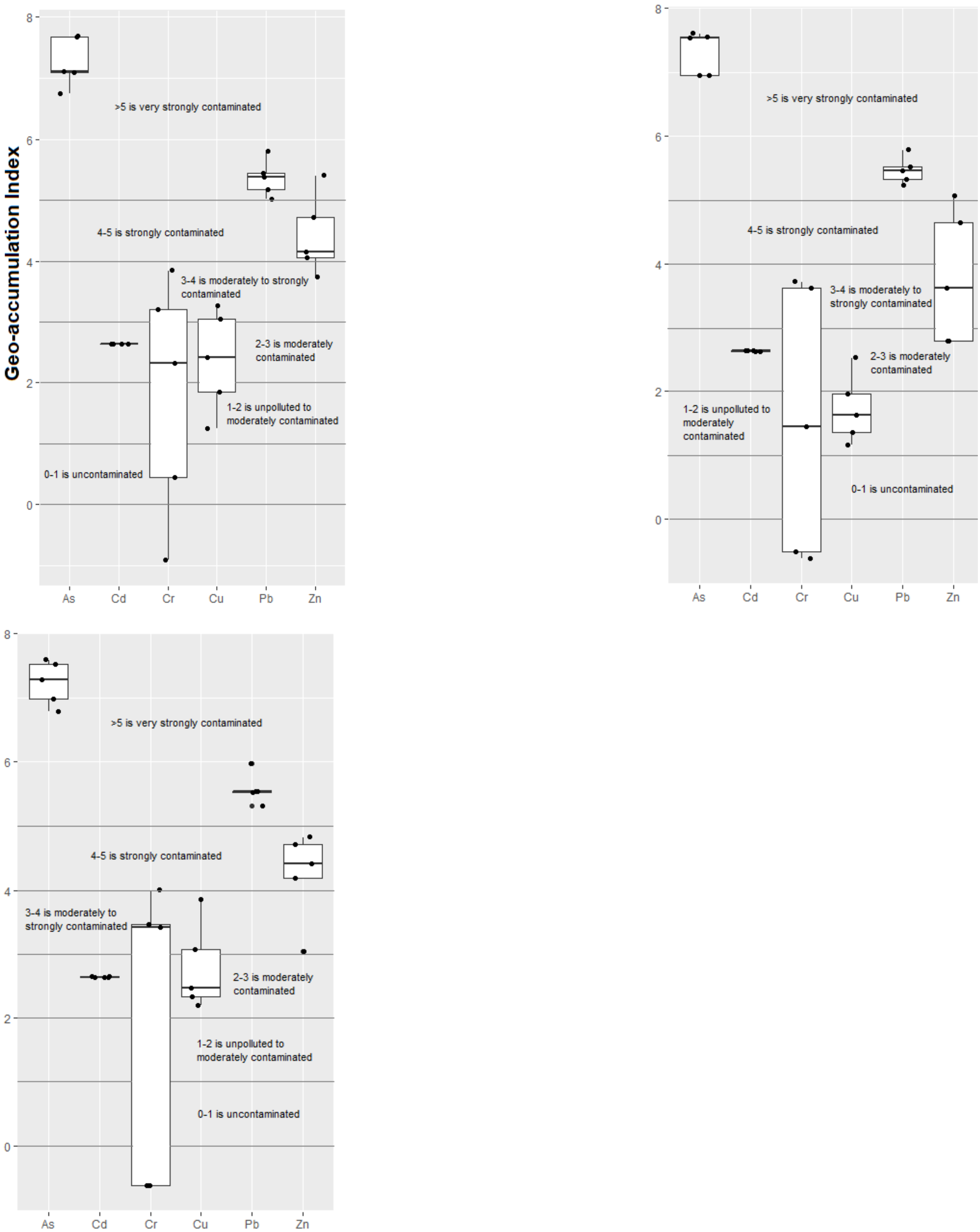


Fig 6. Geo-accumulation Index (Igeo) of heavy metal on post-mining bauxite from different soil level

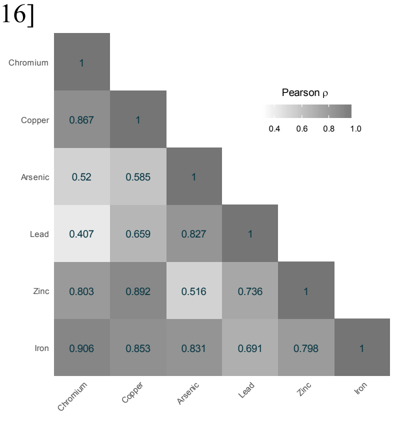
3.4 Correlation Coefficient Matrix (r^2)

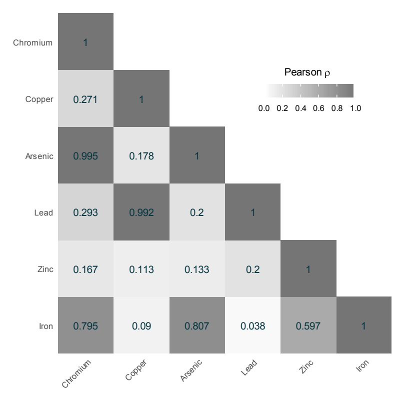
Linear regression analysis showed the strong positive correlation between Fe and every heavy metal concentration (Cr, Cu, As, Pb, Zn) on surface layer post-mining bauxite production (**Fig.7**). The strong significant positive correlation is found in surface layer among Fe and Cr (r^2 = 0.906), Fe and Cu (r^2 = 0.853), Fe and As (r^2 = 0.831), Fe and Pb (r^2 = 0.691), Fe and Zn (r^2 = 0.798). Furthermore the significantly positive correlation from heavy metal concentration in surface layer of Cr (r^2 = 0.803), Cu (r^2 = 0.892), Pb (r^2 = 0.736) with Zn. The toxic pollutant Pb and As have significantly high positive correlation (r^2 = 0.827).

Middle layer had different pattern from surface layer (0-30 cm). Correlation coefficients between pairs of metals show that Cr and As (r^2 = 0.995), Cu and Cd (r^2 = 0.992) that showed the strong positive correlation, others showed of the correlation coefficients that are not at a significant value in middle layer. The values of Pearson´s correlation coefficients that confirmed that bottom layer almost have similar pattern from middle layer. The influence of anthropogenic activities from bauxite production result indicated significant positive correlation among Fe and Cr (r^2 = 0.923), As and Cr (r^2 = 0.772), Cu and Zn (r^2 = 0.951), Pb and Fe (r^2 = 0.823).

Pearson's correlation (PC) matrix for analyzed sediment parameters was calculated to see if some of the heavy metal interrelated with each other [33] and provide information on the sources and pathways of heavy metal pollution [34]. The contamination and high correlations between specific heavy metals in sediment in surface layer, this showed that heavy metal pollutants were most likely derived from same source similar levels of contamination and/or release from the same sources of pollution, mutual dependence and identical behavior during their accumulation in sediment system, may be due to the contamination from production and tailing bauxite production. [12,33].

The results of correlation matrix of each core sample indicate that a significant fraction of the trace metal is found adsorbed on to Fe (surface level), geochemical phases controlling the trace metals in sediments which may be attributed to their large surface area [16]





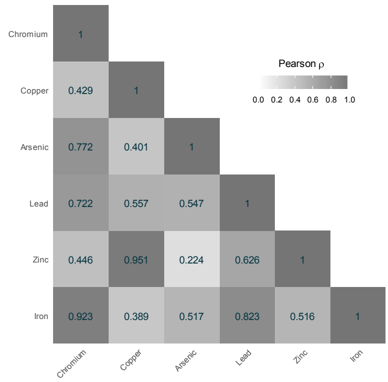


Fig 7. CCM (r^2) for heavy metal concentration in three level core sediment (n=105) from post-bauxite mining

3.5 Effect of heavy metal contamination for aquaculture water quality

Tabel 2. Water quality parameters of post-mining bauxite Bintan Island

Sites	pH	Dissolved Oxygen	Temperature
Sei.Ladi	4.94 ± 0.006	6.43 ± 0.208	30.2 ± 0.115
Sei.Carang	5.56 ± 0.027	6.37 ± 0.513	30.4 ± 0.231
Dompak	4.45 ± 0.058	7.33 ± 0.058	29.7 ± 0.001
Simpang Dompak	6.36 ± 0.046	6.37 ± 0.058	29.1 ± 0.001
Wacopek	6.13 ± 0.057	5.93 ± 0.058	28.7 ± 0.058

The physicochemical parameters of the water column such as pH, temperature and dissolved oxygen are presented in Table 2. The physicochemical parameters are very important because they have significant effect on the water quality [33]. Furthermore, aquatic life also suffer due to degradation of water quality. Among the external factors temperature, pH and dissolved oxygen are important factors which influence the aquaculture ecology.

The average values pH water column post-mining bauxite were ranged from 4.45 with SD ±0.058 to 6.36 with SD ±0.046. Dissolved oxygen (DO) refers to the oxygen gas that is dissolved in the water and made available to aquaculture. The average dissolved oxygen was 6.43 with SD ±0.208 to 7.33 with SD ±0.058. Temperature was measured in water column and the values average of temperature were ranged 28.7 with SD ±0.058 to 30.4 with SD ±0.231.

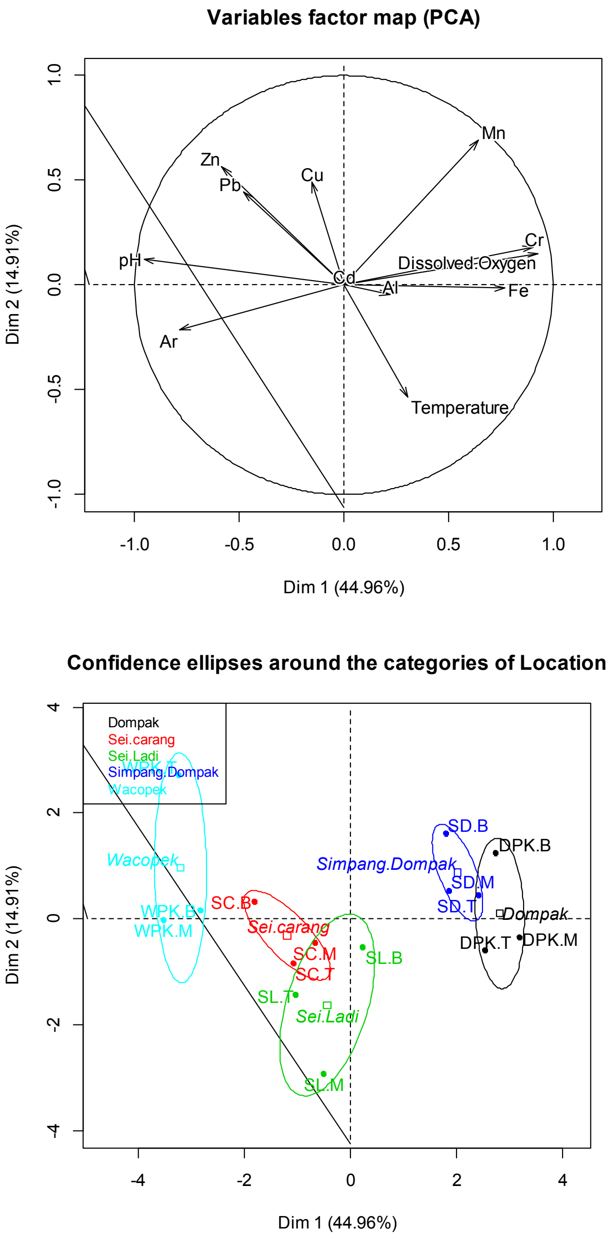


Fig 8. Principle Component Analysis of Effect of heavy metal contamination for water quality post-mining bauxite (a) and confidence ellipse correlation matrix around the categories of site location post-mining bauxite. In both figures the factor loadings of the PCA-axes 1 and 2 are shown, which explain both figure have 44.96% and 14.91% of the variation. Data standardized to mean zero and unit deviance prior to analysis.

The first two axes of the PCA explain 59.87% of the variation between effects of heavy metal contamination for water quality in post-mining bauxite. Dissolved oxygen (DO) were clustered along the first axis with Manganese (Mn) and Chromium (Cr). Temperature was clustered with iron (Fe) and Aluminum (Al) since their arrow on the x-axis pointed in the same direction (Fig 8a). pH was clustered with copper (Cu), Zinc (Zn) and lead (Pb) and was negatively correlated with temperature water column and their exact opposite scores on the x-axis. Dissolved oxygen (DO), Mn and Cr varied independently from temperature and pH, since their lines in the PCA plot are positioned perpendicular to each other (Fig. 8a.) site clustering show similar condition water quality parameter and heavy metal concentration with Dompak and Simpang Dompak in same clustering with intersect on confidence ellipse around (Fig 8b.). Sei Carang and Sei Ladi intersect on confidence ellipse around that show their water quality parameter and heavy metal concentration have similar condition but not significant in the path analysis. Wacopek have different condition from other station

4 CONCLUSIONS

Free-ranging herbivores of interest in study were fishes in the family Acanthuridae (Surgeonfish). Scaridae (parrotfishes). and Siganidae (Rabbitfishes) was found 41 species herbivore fish; Scaridae (25 Species). Siganidae (10 Species) and Acanthuridae (six species). The highest abundance and biomass of herbivorous fish were site RS15 at Penagi on east Natuna. The environmental condition in Natuna Island has good condition with the less of ecological stresses although in several site (at the western Natuna) have been function as destructive fishing area). The post-mining bauxite production in Bintan Island show textural facies of sediment dominated by sand (medium sand and fine Sand). Overall, most of every layer sediment enriched with fine sand and has relatively high sorting values categorized by poorly sorted. Kurtosis analysis show that all sample are better sorted at the tail than the central portions (approximately symmetrical), hence the mesokurtic curves. Grain size distribution of post-bauxite sediment are generally associated with vertical sediment layer. In view of this, the small particles of sediment dominated in surface layer while the larger particles concentration in bottom layer. The sequence of EF for heavy metals in the sediment of post-mining bauxite is in the following order: As > Pb > Zn > Cr > Cu > Cd. This indicates that arsenic was more abundant when compared with the others metals, where cadmium had the lowest appearance. Igeo values showed very strongly contaminated pollution with arsenic (As) and lead (Pb) at every level of soil from post-mining bauxite production. For zinc (Zn), the values of Igeo showed moderate to strongly contaminated pollution degree. On the basis of above said classification, Cr and Cu in both showed uncontaminated to moderately pollution load. The average values pH, dissolved oxygen and temperature water column post-mining bauxite were ranged from 4.45 with SD ±0.058 to 6.36 with SD ±0.046, 6.43 with SD ±0.208 to 7.33 with SD ±0.058 and 28.7 with SD ±0.058 to 30.4 with SD ±0.231 respectively.

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