

# Environmental Impact of Metals Leaching Generated from Long Term Coal Ash Disposal Placement of more than 10 Years Periods

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**ABSTRACT**— *In this paper, we describes the impact of long-term coal ash disposal. Constraints of coal ash waste utilization are the main reason the wastes ended at ash disposal facility for long periods of time generated an environmental impact. The impact is observed based on comparison of several variables such as leaching ratio (LR) values of each environmental component. Leaching potential was completed by calculated LR values and the relationship between the variables through correlation analysis. The largest leaching potential are as follows impact source > waste > disposal > soil. Therefore order of pH as acidity level is reversed when compare to its leaching potential value (pH coal < pH fly ash < pH bottom ash < pH disposal and soil). Furthermore, highest leaching potential viewed as metal concentration are as follows: Sn > Pb > Cr > Mo > Cu > Zn > Li > Co > V > B > Ni > Cd.*

**Keywords**— Disposal, Fly ash, Bottom ash, Coal, Leaching Ratio, Soil, Metal

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## 1. INTRODUCTION

Indonesian government has issued a national energy policy to increase coal usage of at least 30% by 2025. Increase in coal demand is triggered by acceleration program of coal-fired steam power plants construction as consequence of Indonesian ministerial decree on electricity supply in 2016-2025. Coal-fired steam power plants will dominate type of power plant to be built up to capacity of 34.8 GW. The main problem of a coal fuel energy power plant is the solid waste generation in form of bottom ash and fly ash. Indonesian government regulations categorized fly ash and bottom ash from coal combustion processes in steam power plant, boiler and industrial furnace facilities as hazardous and toxic wastes from specific source. According to Hanan et al. (2017) massive production of coal ash has a negative affect to the environment by contaminating underground, adjacent soil and surface water due to limitation of dumping space. Coal ash as off today is still being treated as waste and placed in impoundment ponds, silos or disposals. Activation of heavy metals at dumping location increases the chances of metal release to environment and also strengthens receptor toxicity (Davies 1980). Research conducted by Choi et al.(2002) suggests the disposal of fly ash is considered a potential source of contamination due to the enrichment and surface association of trace elements in the ash particle. During transport, disposal, and storage phases, the residues from coal combustion are subjected to leaching effects of rain and part of the undesirable components in the ashes may pollute both ground and surface waters (Benito et al. 2001). At that point, according to Gupta et al. (2017) coal fly ash is waste material in huge quantity which not only occupies the large area of land, but also causes many environmental issues and effects due to heavy metal leaching.

This study was aimed to determine the environmental impact of long term ash disposal in a period of more than 10 years which took place at two coal-fired steam power plants in Sumatra-Indonesia. Coal supplied to those operations also supplied a steam power plant in West Java-Indonesia, which currently is a subject of an ongoing similar study. Hence, the coal used at Sumatra and West Java operations were originated from the same source, therefore the correlation between contaminants presence in coal source with the magnitude of waste generations can be identified. Analysis and discussion was conducted based on leaching ratio from several variables such as impact source (coal ash), waste (fly ash and bottom ash), waste management at disposal facility, environmental impact analyzed (sub surface soil at disposal area) and environmental impact (leachate water produced by rains and/or run off at leach pond around coal yard and ash disposal area)..

## 2. METHODOLOGIES

### 2.1 Sample Collection

#### a. Sampling location and sample type

Sample collection was conducted at selected locations at Sumatra Indonesia with difficulties in utilizing coal ash waste and has been undertaking long term coal disposal activity as presented in Table 1. (1) Power Plant A: Each unit has two different coal sources: Unit 1 known as coal-1 originated from Sawah Lunto has characteristic of glossy black and high-calorie, while unit 2 known as coal-2 originated from Muaro Bungo Jambi has characteristic of brownish color and lower calorie. (2) Power Plant B: Only one source known as coal-3 originated from Bukit Asam has characteristics of blackish, low calorie, high volatile matter and high moisture however it has low sulfur content and low carbon. Collected samples from ash disposal facility were coal, fly ash, bottom ash, and ash where it has not been utilized and placed at disposal facility since the beginning of the power plant operation. Power plant A was established in 1996 and Power Plant B in 1987. Subsurface soil and leachate water samples were also collected to identify the impact to the environment.

**Table 1:** Type of Sample

Location		Source of Impact	Waste Type		Waste Management	Environmental Impact	
			Processed Ash		Disposed Ash	Leachate	Soil
Power plant-A	Unit-1	Coal-1	Fly ash-1	Bottom ash-1	Disposal-A	Leachate-A	Soil -A
	Unit-2	Coal-2	Fly ash-2	Bottom ash-2			
Power plant-B		Coal-3	Fly ash-3	Bottom ash-3	Disposal-B	Leachate-B	Soil -B

#### b. Sample Collection Methods

Composite samples of coal, disposed ash and suspected contaminated soil were collected after a method conducted by Carter & Gregorich (2008) where ash disposal facility is divided into 10 grids where samples were collected from each grid and composited and mixed in a container. Approximately 300 gr of the composited sample were taken and treated as single sample for total metal content analysis. Fly ash which located in Electrostatic Precipitator (EP) while bottom ash which located under a burning furnace were collected using a heat-resistant container. Composite leachate water samples were collected according to USEPA (2013) by way of combining samples from several leachate locations which were collected at morning, noon and night. The composite leachate water sample were identified and treated as single sample for total metal content analysis.

### 2.2 Analysis Method

#### a. Sample preparation

Samples digestion were performed according to (USEPA, 1996) where strong acid was utilized to extract metals widely presents in nature from coals and disposed ash. Approximately one or two grams of composite homogenized wet samples were prepared and strong acids were added to the sample. The sample was heated until the temperature reached approximately  $95^{\circ}\text{C} \pm 5^{\circ}\text{C}$ .

#### b. Determination of metals using ICP-MS

Total metals determination was conducted through ICP-MS analysis by Thermo Scientific XSERIES 2 Quadrupole. Sample is fed into argon-based high temperature plasma as carrier gas after previously was nebulized. Energy provided by the plasma decomposed, atomized and ionized the sample. Ions generated by energy transfer process was extracted from the plasma through differential vacuum interface and separated based on their atomic mass by mass spectrometer. The operation designs of quadrupole or magnetic sector mass spectrometer made it possible for ions passing through the mass spectrometer to be calculated by electron multiplier detector. The calculation then processed at data processing system.

#### c. Toxicity Characteristic Leaching Procedure (TCLP)

Metal content leaching potential of coal, coal ash and soil was obtained through TCLP test .The TCLP simulation was performed in laboratory according to EPA Method 1311 (1992) and APHA (2012) using acetic acid buffer pH 4.9 (Dungan & Dees 2009) and rotated for  $18 \pm 2$  hours. Exception was made for Mercury TCLP procedure which follows the USEPA (2005).

#### d. Correlation analysis

The pH was measured in situ using soil pH meter, Lutron-pH-212 series. The relationship of pH measurement to TCLP metal concentration was calculated using Pearson Correlation SPSS Statistics, it is used to identify relationship

between two or more variables simultaneously (Rodger & Nicewander, 1988). Suggested Interpretation for Correlation Coefficient by Guilford (1956).

### 3. RESULTS AND DISSCUSSION

#### 3.1 Metals Leaching Ratio

The leaching potential of metals from impact sources, waste and disposal ash is the ratio of TCLP results to total concentration metals. As for soil samples, leaching concentration is not attained from laboratory result simulation but compared with leachate water around the site. As presented in **Table 2** and **Table 3**.

**Table 2:** Metals Leaching Ratio (%) of Power Plant A

No	Parameter	Source of impact		Waste				Management of waste	Impact
		Coal-1	Coal -2	Fly ash 1	Fly ash 2	Bottom ash 1	Bottom ash 2	Disposal-A	Soil-A
1	Ca	0.57	0.64	0.81	0.81	0.97	0.81	1.37	90.73
2	Fe	0.60	1.49	0.88	0.64	0.94	0.64	1.79	0.00
3	Al	0.67	2.22	0.98	0.60	0.85	0.60	0.93	0.01
4	Na	33.32	8.34	14.29	16.67	20.00	16.67	11.11	436.00
5	Mg	1.79	6.66	3.45	2.70	5.88	3.70	4.76	10.96
6	S	1.75	1.85	4.76	2.78	6.25	4.17	4.00	8.27
7	Mn	4.76	3.70	4.17	4.17	4.35	4.35	1.15	0.11
8	K	8.33	6.25	5.56	5.56	6.67	3.85	0.89	4.52
9	Si	4.55	3.23	3.45	3.45	3.57	3.70	0.86	89.30
10	Sr	12.50	20.00	20.00	14.29	12.51	16.67	6.25	44.00
11	Ti	11.11	25.00	16.67	11.11	12.50	12.50	6.25	0.15
12	Cr	50.00	30.00	33.45	33.48	20.00	20.00	10.00	5.00
13	As	8.92	16.15	6.96	5.95	5.75	8.25	5.00	126.67
14	B	20.00	14.29	33.33	20.00	16.66	16.67	4.17	7.50
15	Ba	16.64	16.68	14.29	20.00	14.28	14.29	3.57	0.53
16	Be	10.00	12.50	12.50	14.38	20.00	12.50	2.86	2.00
17	Cd	32.86	34.00	11.11	16.82	12.00	9.55	6.47	0.36
18	Co	34.62	25.20	20.00	12.50	33.33	20.00	5.00	0.11
19	Li	28.00	20.00	33.36	14.29	20.00	25.00	8.40	2.00
20	Mo	33.33	34.00	25.00	16.61	14.12	33.16	5.00	6.67
21	Ni	45.06	11.00	20.00	17.50	12.52	20.00	3.58	0.59
22	Pb	15.34	10.17	10.24	10.17	70.00	71.58	80.32	0.97
23	Sb	16.00	15.00	16.67	33.50	20.00	14.62	5.00	0.00
24	Ag	10.00	12.50	10.67	5.00	1.11	10.00	5.31	3.33
25	Se	10.00	10.00	30.00	20.00	30.00	30.00	6.56	0.82
26	Sn	42.00	50.00	41.00	47.00	48.57	50.00	52.00	0.25
27	V	50.00	20.00	12.49	20.00	11.08	20.00	4.15	0.12
28	Zn	33.33	14.25	33.33	33.33	20.00	25.00	3.57	0.51
29	Hg	7.35	6.67	8.11	5.13	6.00	4.00	3.64	0.04
30	Cu	20.00	22.60	20.00	33.35	33.36	25.00	5.00	6.67

It is obvious that the extractable metal ratios (in terms of leaching ratio calculated as Eq.3.1) (Jiang-shan et al., 2017).

$$\text{Leaching Ratio (LR)} = \frac{\text{Leaching Concentration}}{\text{Total Concentration}} \times 100 \% \quad (\text{Eq. 3.1}).$$

**Table 3 : Metals Leaching Ratio (%) of Power Plant B**

No	Parameter	Source of Impact	Waste		Management of waste	Impact
		Coal-3	FA-3	BA-3	Disposal-B	Soil-B
1	Ca	8.33	1.33	1.54	0.93	0.47
2	Fe	9.09	1.85	2.22	1.28	0.002
3	Al	5.27	0.96	1.03	0.95	0.0005
4	Na	9.10	2.94	2.78	1.02	7.60
5	Mg	14.28	4.35	3.70	1.03	0.13
6	S	6.68	7.69	5.26	3.13	0.95
7	Mn	33.40	11.11	11.11	8.33	0.01
8	K	25.00	5.88	20.00	4.76	21.36
9	Si	20.00	26.92	25.00	12.50	4.97
10	Sr	14.33	20.00	7.14	4.17	0.22
11	Ti	11.23	14.28	9.09	11.11	0.01
12	Cr	30.00	14.09	33.00	20.00	2.00
13	As	20.00	25.00	12.11	7.50	1.17
14	B	12.60	5.27	14.29	20.00	0.04
15	Ba	16.80	16.67	11.11	9.09	0.05
16	Be	33.33	33.33	25.00	20.00	0.14
17	Cd	20.00	20.00	20.00	14.62	0.59
18	Co	37.50	50.00	12.78	11.14	0.02
19	Li	25.00	33.29	16.63	25.00	0.40
20	Mo	50.00	25.00	20.00	20.00	0.53
21	Ni	22.22	20.00	25.00	25.26	0.11
22	Pb	50.00	18.39	11.00	11.14	0.09
23	Sb	33.33	15.00	35.00	10.00	0.77
24	Ag	26.67	20.00	13.33	20.00	7.69
25	Se	25.00	33.33	15.00	26.92	0.42
26	Sn	25.22	33.00	17.00	12.67	0.50
27	V	16.80	20.00	25.00	20.00	0.02
28	Zn	20.00	16.71	14.33	33.36	0.01
29	Hg	1.11	15.79	1.11	6.41	0.08
30	Cu	28.00	25.17	10.00	33.40	0.02

The LR value for parameters Ca, Na, As, Sr and Si in soil samples of Power Plant A increased sharply compared to the one on impact sources, waste or ash disposal. The value shows that the elevated result affected by natural occurrence of the soil. However, the result show different trend at Power Plant B where LR value is very low except for Potassium (K) which amounted up to 21.36%.

The largest leaching potential in the form of LR of metals from impact source, waste, waste management and environmental impacts shows comparison as follows coal > bottom ash > fly ash disposal > soil. The highest LR can be found at the impacts source compared to the waste and environmental impacts.

LR value of metals found at Power Plant A and B are as Sn > Pb > Cr > Mo > Cu > Zn > Li > Co > V > B > Ni > Cd. As shown on Figure 1, the parameters generated from coal, ash and soil were generally have stable LR, except for Pb that occurred in highest percentage after converted as bottom ash and disposed to designated temporary storage. This has been reported by Baba & Kaya (2004), the results of the geochemical analyses of Pb concentrations of these toxic trace elements are higher than in the original coal. The enrichment of elements in fly ash was observed to be higher than that of bottom ash.

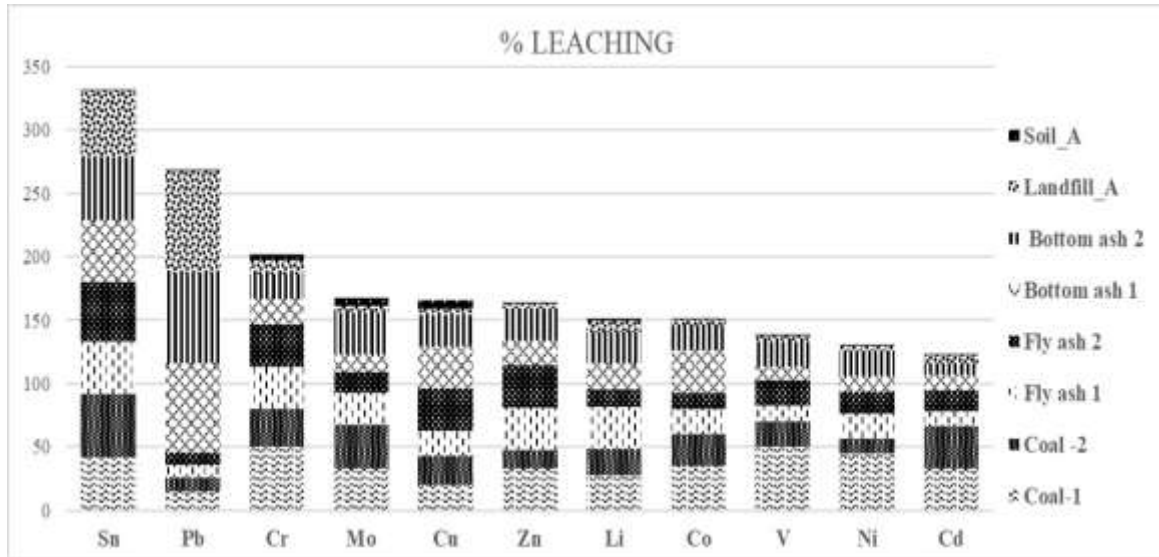


Figure 1: Metals with Highest Leaching Ratio

### 3.2 . Correlation with pH

According to Lau & Wong (2001) different elements have different leaching behaviors due to different elemental properties and 'solution pH' as well as leaching time, which greatly affects leaching. Based on in situ pH measurement, the most acidic pH occurred in impact source coal and increases as the ash is disposed over time as shown in Figure 2. However, pH of the surrounding soil at disposal area is similar to it of disposed ash. Significant correlation of <0.05 is determined using bivariate correlation of pH measurement and compared them to the laboratory result. The significant pH correlation is showed on metal content result such as Pb, Cd, Al, Li, Se and S (Table 4).

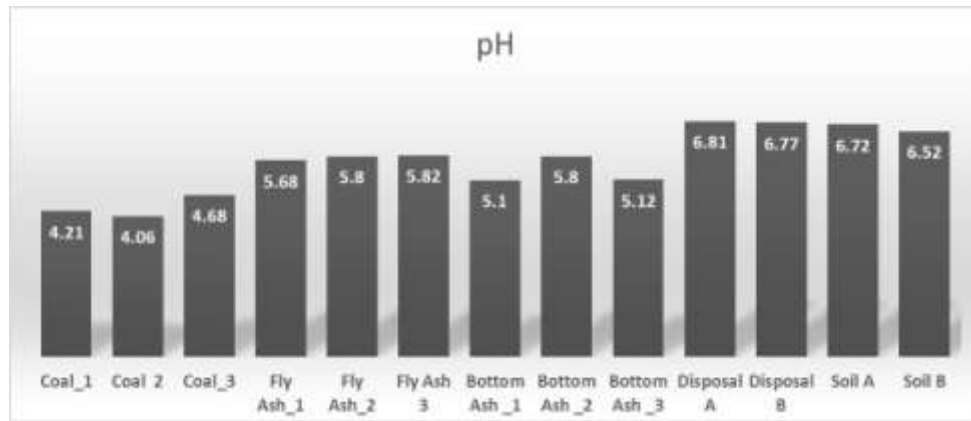


Figure 2 : Result of pH measurement

Table 4: The Significant pH Correlation

Pb	Pearson Correlation	0.580*	Cd	Pearson Correlation	0.519*
	Sig. (2-tailed)	0.023		Sig. (2-tailed)	0.048
Al	Pearson Correlation	0.540*	Li	Pearson Correlation	0.515*
	Sig. (2-tailed)	0.038		Sig. (2-tailed)	0.05
Se	Pearson Correlation	0.533*	S	Pearson Correlation	-0.572*
	Sig. (2-tailed)	0.041		Sig. (2-tailed)	0.026

\*correlation is significant at the 0.05 level (2-tailed)

Aluminum content which correlated with pH is in line with the study performed by Wright et al. (2007), pH of stream water is an important control of trace metal solubility where pH values are low, dissolved aluminum concentrations generally are high; where pH values are high aluminum concentrations are low. On the other hand, TCLP results of Al do not show a large percentage. Haiyan et al. (2013) concluded that higher leaching rates occur at low pH, with leaching sequence being  $Zn \geq Cu > Pb > Cr > Cd$ , but this study results show the leaching sequence as follows  $Pb > Cr > Cu > Zn > Cd$ . Although pH is the most important soil property in determining the retention of the free metal ion, distribution coefficient ( $K_d$ ) values based on total dissolved metal in solution may show little pH dependence for metal ions that have strong affinity for dissolved organic matter (Degryse et al., 2009). Potential leaching on coal > bottom ash > fly ash > disposal > soil compared to pH value (Figure 2) where pH Coal < pH Fly ash < pH decreases with increasing bottom ash < pH Disposal and Soil. This suggests that the greater the pH the leaching rate is decreases as written on Gould et al. (1989) in which metal solubility generally decreases with increasing pH.

### 3.3 Environmental impact of long term coal ash disposal

The principle of environmental impact of coal ash management through disposal is representation of comparative study of long term disposal activity of more than 10 years at disposal facility operated at Power Plant-A < Power Plant-B. The study showed no significant impact on the surrounding soil and water quality. Moreover leaching percentage of soil-B, is lower than in soil, whereas duration of disposal-B > disposal-A. However, it is known that leaching potential at disposal-B is greater than 65.44% compared to disposal-A as presented in Figure 3. This shows even though leaching potential in disposal site is higher, the impact to environment is not necessarily increased due to leaching ratio disposal-B is lower than disposal-A. This can be influenced by natural factors such as original soil quality and rainfall rate as explain by Naupene et al. (2017) which stated that based on this rate, controlled leaching by diffusional process responsible for transferring these elements from interior to the surface of the particles as well as the dissolution of the fly ash particles. According to Singh et al. (2012) since the soil below the impoundments is always saturated and under considerable hydraulic head, the inefficiently lined ponds provide a great opportunity for groundwater contamination to seep in. Therefore, the seepage from ash pond may be more as compared to leaching from landfills and ash mounds. But for the future, it is important to adopt effective containment/treatment schemes to avoid potential and persistent dispersion of trace elements from ash disposal facilities to surrounding environment for a long time (Naupene et al., 2017).



Figure 3: Potential leaching of Disposal Power Plant A and Disposal Power Plant B

## 4. CONCLUSIONS

Coal ash waste management by means of final disposal at ash disposal area in long term is thought to increase potential leaching resulting in a decrease in soil and water quality. The results of this study can be concluded that the disposal as environment management of 'end of pipe' when no more solution to manage coal ash can still be done. But with proper location selection requirements take into account pH of rain, rainfall, rain intensity, soil physics-chemical condition, topography and local geology. Besides, the disposal sites are designed to avoid leachate in rainy season and coal ash pile is not in direct contact with soil.

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