Utilization of Coal Bottom Ash and Cattle Manure as Soil Ameliorant on Acid Soil and Its Effect on Heavy Metal Content in Mustard (Brassica juncea)

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ABSTRACT

Coal bottom ash and cattle manure can be used as soil ameliorant. The application of coal bottom ash and cattle manure can improve the soil chemical properties, such as pH and the amounts of available nutrients in soil. The objective of the study was to understand the effect of coal bottom ash and cow manure application on soil chemical properties and heavy metal contents in soil and mustard (Brassica juncea). A pot experiment was conducted in a greenhouse, including three treatment factors, i.e. age of coal bottom ash (fresh, 4 months and 2 years), dose of coal bottom ash, i.e. 0, 40 and 80 Mg ha⁻¹, and dose of cattle manure, i.e. 0 and 10 Mg ha⁻¹. The results show that the application of coal bottom ash and cattle manure increased the pH and the amounts of total-N, available-P and exchangeable cations (K, Ca and Mg) of the soil. The application of coal bottom ash increased the amounts of Pb, Cd and Co in the soil, but did not increase the amounts of Pb and Co in mustard, while the application of cattle manure increased the amount of Cd both in soil and mustard.

Keywords: Coal bottom ash, cattle manure, heavy metal, mustard, soil ameliorant

INTRODUCTION

Coal ash is the waste of coal combustion. Based on its particle size, coal ash consists of fly ash and bottom ash. Coal combustion produces about 5% ash that consists of 80% up to 90% fly ash and 10% up to 20% bottom ash. According to the data from Indonesian Power Plant, approximately 2 million Mg of coal ash was produced in 2006 (Aziz et al. 2006). The amount of coal ash produced is
continuously increasing every year, resulting in the accumulation of coal ash in the landfill.

The utilization of coal ash in Indonesia is regulated by the Government Regulation Number 101/2014, which classifies coal ash, either fly ash or bottom ash, as one of B3 waste (hazardous and toxic substances), thus in its utilization must be tested by The Toxicity Characteristic Leaching Procedure (TCLP). The regulation challenges the utilization of coal ash as soil ameliorant. On the other hand, the study on coal ash as soil ameliorant has already performed. The fly ash is used more often than bottom ash as soil ameliorant in the recent studies of the coal ash utilization because the fly ash has smaller size, \textit{i.e.} \(0.001 – 100\ \mu\text{m}\) (Haynes 2009), compared to bottom ash \((0.1 – 10\ \text{mm})\) (Korcak 1995). In addition, fly ash has high porosity that contributes to the cation leaching.

According to Korcak (1995), coal bottom ash contains K, Ca, Mg and Na that are essential nutrients for plants. In addition, the study conducted in the greenhouses and experimental fields by Sell et al. (1989) suggests that coal bottom ash is potential to be used as a soil additive that will not damage the soil, crops, or environment.

Another potential soil ameliorant is organic matter. Organic matter can increase cation exchange capacity (CEC), affect pH and increase availability of nutrients, as well as the source of energy for microorganisms in soil (Stevenson 1982).

Utilization of coal bottom ash on acid mineral soils is potential to be developed to increase the soil productivity. Inceptisol is the soil type with relatively low fertility status, acidic pH (pH 4.5) and low up to medium base saturation (Sudirja et al. 2007). Inceptisols occupy approximately 40% or 70.52 millions ha of the total area of land in Indonesia (Puslitbangtanak 2003) and can be attempted for the expansion of agricultural land. In Indonesia, mustard (\textit{Brassica juncea}) is widely cultivated and consumed as vegetable. However, mustard is a hyperaccumulator plant for heavy metals.

The study was conducted to evaluate the feasibility of coal bottom ash as a soil ameliorant, to study the effect of coal bottom ash and cow manure application on the soil chemical properties of an acid mineral soil and to study the effect of coal bottom ash and cow manure application on the heavy metal content in soil and mustard.

**MATERIALS AND METHODS**

The materials used in this study was the coal bottom ash obtained from the landfill of Power Plant PLTU Paiton, soil sample of Inceptisol taken from Dramaga, Bogor, cattle manure, and Urea fertilizer.

A pot experiment was conducted in a greenhouse using a Completely Randomized Design (CRD) with three factors and three replications. The first factor was the age of coal bottom ash (fresh, 4 months and 2 years); the second factor was the dose of coal bottom ash \((0, 60\text{ and }120\ \text{g pot}^{-1}, \text{equivalent to }0, 40\text{ and }80\ \text{Mg ha}^{-1})\); and the third factor was the dose of cattle manure \((0\text{ and }15\ \text{g pot}^{-1}, \text{equivalent to }0\text{ and }10\ \text{Mg ha}^{-1})\), so overall there were 54 experimental pots. The amount of soil used in the pot experiment was 3 kg of air dried soil/pot. Coal bottom ash and cattle manure were mixed homogeneously with the soil and incubated for 7 days. Then, the mustard seeds were planted. Urea fertilizer was applied 2 weeks after planting. Four weeks after planting, the soil and plant samples were taken for analysis.

The soil properties measured were pH (pH-meter), total-N (Kjeldahl), available-P (Bray 1), and exchangeable-K, -Ca, and -Mg \((\text{NH}_4\text{OAc} 1\text{N pH} 7\) extraction, K measured using flame photometer, Ca and Mg measured using AAS). In addition, the heavy metal content (Pb, Cd and Co) was measured in the soil and plant samples \((\text{HClO}_4\text{ and }\text{HNO}_3\) destruction, measured using AAS). The data were statistically analysed using Analysis of Variance (ANOVA) and further tested using Duncan’s Multiple Range Test (DMRT) at 5% significance level.

**RESULTS AND DISCUSSION**

**Characteristics of Coal Bottom Ash and Cattle Manure**

Fresh coal bottom ash was taken directly from the silo. The chemical characteristics of fresh coal bottom ash are in general similar to the characteristics of coal bottom ash that has been piled for 4 months and for 2 years in the landfills (Table 1). This result is in contrast with the study of Iskandar et al. (2013), which indicated that the longer the fly ash dumped in the landfill, the lower its pH and the exchangeable cation content are. This is probably related to the particle size of the coal bottom ash that is bigger than the coal fly ash, so it would not be affected by the high levels of leaching. Haynes (2009) also stated that the type of coal used during the combustion process determines the chemical characteristics of coal bottom ash.

The use of coal bottom ash can create a problem for environment due to its heavy metal content, so the utilization of coal ash is restricted by
the Government Regulation Number 101/2014. The concentrations of heavy metals Pb, Cd, Co, Cr, Ni, As and Hg in the coal bottom ash used in this study are in the normal range for heavy metal content in soil. The highest concentrations of Pb, Cd, Co, Cr and Ni are measured in the 2 years age coal bottom ash, but in comparison with the data from Alloway (1995) these levels are classified as normal concentrations of heavy metals in soil. The total concentrations of heavy metals in coal bottom ash are presented in Table 2.

Cattle manure is one of the organic materials that is widely used in composting. The purpose of composting is to decompose the fresh organic materials into substances like humus (Indranada 1986). In the process of composting, the organic materials change bio-physically and chemically involving the activity of microbes and mesofauna. Suryadikarta and Simanungkalit (2006) indicated that compost contains all nutrients in various amounts depending on the type and origin of materials of

Table 1. Chemical characteristics of coal bottom ash used in the study.

| Characteristic   | Age of Coal Bottom Ash |
|-----------------|------------------------|
|                 | Fresh | 4 months | 2 years |
| pH H₂O          | 6.60  | 6.90     | 6.60    |
| SiO₂ (%)        | 57.40 | 55.50    | 57.8    |
| Al₂O₃ (%)       | 17.57 | 18.09    | 13.79   |
| Fe₂O₃ (%)       | 13.15 | 13.97    | 16.93   |
| K₂O (%)         | 1.25  | 1.24     | 1.38    |
| Na₂O (%)        | 0.49  | 0.46     | 0.26    |
| CaO (%)         | 5.66  | 4.76     | 4.83    |
| MgO (%)         | 2.67  | 3.57     | 2.43    |
| TiO₂ (%)        | 1.13  | 1.21     | 1.03    |
| MnO (%)         | 0.063 | 0.064    | 0.10    |
| P₂O₅ (%)        | 0.039 | 0.058    | 0.026   |
| LoI (%)         | 0.030 | 0.050    | 0.17    |

Table 2. Total concentration of heavy metals in coal bottom ash.

| Total Metal* | Age of Coal Bottom Ash |
|--------------|------------------------|
|              | Fresh | 4 months | 2 years |
| Pb (ppm)     | 35    | 27       | 61      |
| Cd (ppm)     | 3     | 4        | 4       |
| Co (ppm)     | 26    | 18       | 39      |
| Cr (ppm)     | nd    | nd       | 5       |
| Ni (ppm)     | 21    | 28       | 37      |
| As (ppm)     | nd    | 13       | 11      |
| Hg (ppm)     | nd    | nd       | nd      |

Notes: *Measured using Furnace Atomic Absorption Spectrophotometer
nd = not detected

Table 3. Characteristics of cattle manure.

| Characteristic | Value |
|----------------|-------|
| Water content (%) | 23.74 |
| Carbon content (%) | 12.80 |
| Nitrogen content (%) | 1.53 |
| C/N ratio | 8 |
| Total-P₂O₅ (%) | 0.78 |
| Total-K₂O (%) | 1.13 |
compost, and compost provides nutrients in slow release and limited quantities with the main function to improve soil fertility. The characteristics of cattle manure used in this study are presented in Table 3.

The Effect of Coal Bottom Ash and Cattle Manure Application on pH and Nutrient Content of Soil

The effects of age of coal bottom ash, dose of coal bottom ash and dose of cattle manure on soil pH are presented in Table 4. The results of statistical analysis showed that the doses of coal bottom ash and cattle manure applied on the soil sample significantly increased the soil pH. Meanwhile, the application of different age of coal bottom ash did not affect the soil pH. This is presumably because the pH of fresh bottom ash, 4 months age bottom ash and 2 years age bottom ash are in the same range, which is about 6.60 to 6.90 (Table 1), so that the different age of coal bottom ash applied did not contribute to the increase of soil pH. The study conducted by Oklima (2014) showed that the application of coal ash increased the soil pH.

The effect of combination of age of coal bottom ash, coal bottom ash dose and cattle manure dose on the amount of total-N in soil is presented in Figure 1. The results of statistical analysis showed that the coal bottom ash age, coal bottom ash dose and cow manure dose significantly affected the amount of total-N in soil, and there was an interaction effect of the three treatments. The highest amount of total-N in the soil after application of coal bottom ash and cattle manure was 0.23%. However, based on the criteria of soil characteristics proposed by Balai Penelitian Tanah (2012), the total-N content in the soil samples applied with coal bottom ash and cattle manure measured in this study is in the category of low.

The increased amount of total nitrogen in soil after application of coal bottom ash and cattle manure was probably resulted from the addition of cattle manure, but because the dose of cattle manure applied was low, i.e. 10 Mg ha⁻¹, then the increase of total nitrogen in soil was not significant. Meanwhile, the addition of coal bottom ash did not contribute nitrogen into the soil, because during the combustion process of coal, the nitrogen in coal

| Treatment | pH   |
|-----------|------|
| Age of bottom ash |      |
| T0 (fresh) | 4.36 a |
| T1 (4 months) | 4.30 a |
| T2 (2 years) | 4.37 a |
| Dose of bottom ash |      |
| A0 (0 Mg ha⁻¹) | 4.25 b |
| A1 (40 Mg ha⁻¹) | 4.31 b |
| A2 (80 Mg ha⁻¹) | 4.47 a |
| Dose of cattle manure |      |
| K0 (0 Mg ha⁻¹) | 4.29 b |
| K1 (10 Mg ha⁻¹) | 4.39 a |

T0 : fresh; T1 : 4 months; T2 : 2 years; A0 : 0 Mg ha⁻¹; A1 : 40 Mg ha⁻¹; A2 : 80 Mg ha⁻¹; K0 : 0 Mg ha⁻¹; K1 : 10 Mg ha⁻¹.

Figure 1. The effect of the combination of age of coal bottom ash, coal bottom ash dose and cattle manure dose on the amount of total-N in soil. The same letters above the bar chart indicate no significant difference at 5% significance level according to DMRT. T0 : fresh; T1 : 4 months; T2 : 2 years; A0 : 0 Mg ha⁻¹; A1 : 40 Mg ha⁻¹; A2 : 80 Mg ha⁻¹; K0 : 0 Mg ha⁻¹; K1 : 10 Mg ha⁻¹.
would be lost, so that the amount of nitrogen in the bottom ash is very little or even negligible (Bradshaw and Chadwick 1980).

The effects of age of coal bottom ash, coal bottom ash dose and cattle manure dose on the amount of available-P in soil are presented in Table 5. The results showed that the application of coal bottom ash or cattle manure increased the amount of available-P in soil. Phosphorus is an essential macronutrient needed for plant growth. Inceptisol soils contain low amount of silica, Al and Fe. Al and Fe can bind phosphate in the forms of Al-P and Fe-P, resulting in the decrease of available-P in the soil. A study conducted by Shen et al. (2007) indicates that the phosphorus content in the coal ash is more available in soil, so it is more easily absorbed by plants. Meanwhile, organic matter particularly animal manure can decrease the P fixation by Al and Fe, thereby increasing the availability of P in soil (Suharyani et al. 2012).

The effect of age of coal bottom ash, coal bottom ash dose and cattle manure dose on the amount of exchangeable-K in soil is presented in Figure 2. The application of different age of coal bottom ash, coal bottom ash dose and cattle manure dose showed a significant effect on the amount of exchangeable-K in soil and there was an interaction effect of the three treatments. The application of 2 years age coal bottom ash at 40 Mg ha$^{-1}$ and cattle manure at 10 Mg ha$^{-1}$ (T2 A1 K1) increased the amount of exchangeable-K in soil from 0.40 cmol(+) kg$^{-1}$ (no ameliorant applied/T0 A0 K0) to 1.32 cmol(+) kg$^{-1}$, while the application of 10 Mg ha$^{-1}$ cattle manure without bottom ash (T2 A0 K1) increased the amount of exchangeable-K to 1.35 cmol(+) kg$^{-1}$.

The effect of age of coal bottom ash, coal bottom ash dose and cattle manure dose on the amount of exchangeable-Ca in soil is presented in Figure 3. The results of statistical analysis showed that the combination of age of coal bottom ash, coal bottom ash dose and cattle manure dose significantly affected the amount of exchangeable-Ca in soil, and there was an interaction effect of the three treatments. The application of 2 years age coal bottom ash at 80 Mg ha$^{-1}$ and cattle manure at 10 Mg ha$^{-1}$ (T2 A2 K1) resulted in the highest amount of exchangeable-Ca in soil. In addition, the application of 2 years age coal bottom ash at 40 Mg ha$^{-1}$ and cattle manure at 10 Mg ha$^{-1}$ (T2 A1 K1) also increased the amount of exchangeable-Ca in soil in comparison to other treatments. The amount of exchangeable-Ca in the T2 A2 K1 treatment increased into 6.44 cmol(+) kg$^{-1}$ in comparison to the amount of exchangeable-Ca in the control treatment (T0 A0 K0), i.e. 2.37 cmol(+) kg$^{-1}$, while

![Figure 2](image-url)
The amount of exchangeable-Ca in the T2 A1 K1 treatment increased into 5.08 cmol(+) kg\(^{-1}\).

The effect of age of coal bottom ash, coal bottom ash dose and cattle manure dose on the amount of exchangeable-Ca in soil is presented in Figure 3. The same letters above the bar chart indicate no significant difference at 5% significance level according to DMRT. T0 : fresh; T1 : 4 months; T2 : 2 years; A0 : 0 Mg ha\(^{-1}\); A1 : 40 Mg ha\(^{-1}\); A2 : 80 Mg ha\(^{-1}\); K0 : 0 Mg ha\(^{-1}\); K1 : 10 Mg ha\(^{-1}\).

The effect of age of coal bottom ash, coal bottom ash dose and cattle manure dose on the amount of exchangeable-Mg in soil is presented in Figure 4. The Figure 4 showed a significant effect of the combination of age of coal bottom ash, coal bottom ash dose and cattle manure dose on the amount of exchangeable-Mg in soil, and there was an interaction effect of the three treatments. The addition of 2 years age coal bottom ash at 80 Mg ha\(^{-1}\) and cattle manure at 10 Mg ha\(^{-1}\) (T2 A2 K1) increased the amount of exchangeable-Mg to 1.33 cmol(+) kg\(^{-1}\).

The Effect of Coal Bottom Ash and Cow Manure Application on Heavy Metal Content in Soil

The heavy metal content measured in the coal bottom ash is presented in Table 2. The content of heavy metal selected for this study, i.e. Lead (Pb), Cadmium (Cd) and Cobalt (Co) exceeds the limits of heavy metals in soil based on the criteria proposed by Alloway (1995).

The effects of age of coal bottom ash, coal bottom ash dose and cattle manure dose on the amounts of Pb, Cd and Co in soil are presented in Table 6. The results of statistical analysis indicated that the application of coal bottom ash shows an...
The effects of age of coal bottom ash, dose of coal bottom ash and dose of cattle manure on the amounts of Pb, Cd and Co in soil.

Table 6. The effects of age of coal bottom ash, dose of coal bottom ash and dose of cattle manure on the amounts of Pb, Cd and Co in soil.

| Treatment               | Pb  | Cd   | Co  |
|-------------------------|-----|------|-----|
| **Age of bottom ash**   |     |      |     |
| T0                      | 0.27 a | 0.005 b | 0.28 b |
| T1                      | 0.27 a | 0.066 a | 0.28 b |
| T2                      | 0.27 a | 0.007 a | 0.29 a |
| **Dose of bottom ash**  |     |      |     |
| A0                      | 0.28 a | 0.005 b | 0.29 a |
| A1                      | 0.27 ab | 0.006 b | 0.28 ab |
| A2                      | 0.26 b | 0.007 a | 0.28 b |
| **Dose of cattle manure** |   |      |     |
| K0                      | 0.27 a | 0.006 b | 0.28 a |
| K1                      | 0.27 a | 0.007 a | 0.28 a |

Table 7. The effects of age of bottom ash, dose of bottom ash and dose of cattle manure on the amounts of Pb, Cd and Co in mustard.

| Treatment               | Pb  | Cd   | Co  |
|-------------------------|-----|------|-----|
| **Age of bottom ash**   |     |      |     |
| T0                      | 0.0008 a | 0.0002 b | 0.0012 a |
| T1                      | 0.0009 a | 0.0003 b | 0.0012 a |
| T2                      | 0.0009 a | 0.0004 a | 0.0010 b |
| **Dose of bottom ash**  |     |      |     |
| A0                      | 0.0007 a | 0.0002 b | 0.0011 a |
| A1                      | 0.0009 a | 0.0003 ab | 0.0011 a |
| A2                      | 0.0009 a | 0.0004 a | 0.0012 a |
| **Dose of cattle manure** |   |      |     |
| K0                      | 0.0008 a | 0.0002 b | 0.0010 b |
| K1                      | 0.0009 a | 0.0004 a | 0.0013 a |

The effect on the increasing amount of Pb in soil (Table 6). The amount of Pb measured is in the range of 0.26 to 0.28 ppm, which is still under the normal limit of Pb allowed in soil. According to Alloway (1995), the normal concentration of Pb in soil is about 2-300 ppm.

Table 6 showed that the application of different age of coal bottom ash, coal bottom ash dose and cattle manure dose significantly affected the amount of Cd in soil. The amount of Cd measured in soil ranged from 0.005 to 0.007 ppm. As a comparison, the data from Alloway (1995) indicated that the normal range of Cd in soil is 0.001 to 2 ppm, while the critical limit of Cd in soil is 3-8 ppm.

Table 6 also described the effect of age of coal bottom ash, coal bottom ash dose and cattle manure dose to the amount of Co in soil. The results of statistical analysis showed that the three treatments significantly affected the amount of Co in soil. The highest amount of Co measured is on average 0.29 ppm. Alloway (1995) suggests that the normal range of Co in soil is about 0.5 to 65 ppm.

The Effect of Coal Bottom Ash and Cow Manure Application on Heavy Metal Content in Mustard

The effects of age of coal bottom ash, dose of coal bottom ash and dose of cattle manure on the
heavy metal content (Pb, Cd and Co) in mustard are presented in Table 7. The results of statistical analysis showed that the application of different age of coal bottom ash, coal bottom ash dose and cattle manure dose showed no significant effect on the amount of Pb in mustard. The amount of Pb in mustard leaves after application of coal bottom ash at 80 Mg ha$^{-1}$ in different age or cow manure at 10 Mg ha$^{-1}$ increased on average 0.0009 ppm in comparison to the amount of Pb in mustard leaves without coal bottom ash or cattle manure application (on average 0.0007 ppm). Alloway (1995) suggested that the normal limit of Pb content in plants is about 0.2-20 ppm.

Table 7 shows the significant effect of application of different age of coal bottom ash or cattle manure dose on the amount of Cd in mustard. Alloway (1995) indicates that the normal limit of Cd content in plant is around 0.1 to 2.4 ppm. The highest Cd content in mustard applied with coal bottom ash or cattle manure measured in this study was on average 0.0004 ppm, in comparison to that in mustard without coal bottom ash or cattle manure application (on average 0.0002 ppm). The amount of Cd in mustard measured in this study is in general lower than the allowed threshold level of Cd in edible crops.

The results of statistical analysis indicated that the application of different age of coal bottom ash or dose of cattle manure shows an impact on the increasing amount of Co in mustard (Table 7). The application of fresh bottom ash at 80 Mg ha$^{-1}$ or cattle manure at 10 Mg ha$^{-1}$ resulted in the highest Co content in mustard, i.e. 0.0013 ppm, while the Co content in the mustard without coal bottom ash or cattle manure application resulted in the lowest content of Co, i.e. 0.0010 ppm. Alloway (1995) indicates that the normal content of Co in plants is about 0.1-2.4 ppm, whereas the critical limit is about 4-200 ppm.

**CONCLUSIONS**

The application of different age of coal bottom ash and the dose of coal bottom ash increased pH and the amounts of exchangeable cations (K, Ca and Mg) of the Inceptisol soil. Meanwhile, the application of cattle manure contributed to the increase of total-N and available-P in the soil.

The addition of coal bottom ash at 40 and 80 Mg ha$^{-1}$ increased the amounts of Pb, Cd and Co in the soil, but did not increase the amounts of Pb and Co in mustard. The application of cattle manure increased the amount of Cd in the soil. However, in all treatments, the heavy metal contents measured in the soil are considered low and below the normal limits for heavy metal content in soil.

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