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ORIGINAL PAPER

Different Effects of Ash Application on the Carbon Mineralization and Microbial Biomass Carbon of Reclaimed Mining Soils

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Abstract

Ash resulting from biomass energy resource utilization contains a wide range of metal oxides and hydroxides, which may influence the capacity of the ash to be used as a soil amelioration material. This study aimed to assess the effects of different ashes on changes in soil carbon (C) mineralization and soil microbial biomass carbon (MBC) in reclaimed mining soils (RMSs). Different levels (0, 25, 50, and 75 Mg ha^{-1}) of three ashes (rice husk, oil palm shell, and coal fly ash) were applied to 10-year RMS for a 120-day incubation period. Carbon mineralization was measured over the 120-day incubation period, while MBC and selected chemical properties were quantified at the end of the incubation period. The results of the study showed that the application of rice husk and oil palm shell ash at all levels and coal fly ash at low levels ($\leq$ 25 Mg ha^{-1}) increased C mineralization and MBC. However, the C mineralization and MBC of the soil decreased significantly when the amount of added coal fly ash reached 75 Mg ha^{-1}. These decreases in C mineralization and MBC may be ascribed to the harmful effect of high amounts of coal fly ash on microbial activity and the increased specific surface areas and contents of Ca, Mg, oxalate- and dithionite-extractable iron and aluminum in soil with high amounts of added coal fly ash. This study demonstrates that the application of different types of ash to RMS leads to different C mineralization and soil MBC responses.

Keywords Organic carbon stabilization · Decomposition · Iron oxide · Aluminum oxide

1 Introduction

Several attempts have been made to replace fossil fuels with renewable energy resources, such as biomass energy sources, in response to concerns about climate change (Bentsen and Felby 2012; Varlas et al. 2017). A result of the use of biomass for energy generation is the production of large amounts of ash during incineration. Ash is frequently considered an unwanted product because of its toxic elements, such as Cd, Ni, Pb, Cr, Zn, Co, and Cu (Maresca et al. 2018; Munda et al. 2016; Noyce et al. 2016); therefore, large quantities of generated ash are regularly applied for landfills (Careddu et al. 2015; Valentim et al. 2019). Ash also contains major nutrients required by plants, except for nitrogen, and has liming properties due to its high contents of metal oxides and hydroxides (Maresca et al. 2019; Qin et al. 2017; Silva et al. 2019). Therefore, ash is frequently applied as an ameliorant material to soils to improve soil quality.

Microorganisms are essential soil ecosystem drivers that conduct soil biochemical processes, such as the decomposition of organic matter (OM), nutrient cycling and the production of greenhouse gasses (Paul 2014). Changes in soil ecosystems may affect soil microbial communities, thus eventually influencing soil quality and soil productivity. Carbon mineralization and microbial biomass carbon (MBC) are the most broadly applied variables for measuring the effects of alterations in soil ecosystems on soil microbial processes (Chen et al. 2018; Morillas et al. 2017; Singh and Gupta 2018; Zhao et al. 2019). C mineralization and MBC have been used to describe the effect of long-term nitrogen fertilizer application (de Andrade et al. 2019) and the effect of corn stover management (Urra et al. 2018) on changes in soil quality. MBC is frequently thought to be a more dynamic indicator than those based on physicochemical soil characteristics and therefore has the advantage of being an early warning parameter of changes in soil quality (Bünemann et al. 2018; Schloter

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et al. 2018). The results of these studies suggest that C mineralization and MBC have crucial functions in changes in soil quality throughout the acceleration of soil organic carbon decomposition.

Reclaimed mining soils (RMSs) have an irregular soil layer due to the mining process, so the characteristics of RMSs are very different from those of the original soils. RMSs generally have a low level of soil fertility, with low contents of OM and nutrients, a low soil pH, high contents of toxic elements (Ahirwal et al. 2017; Feng et al. 2019; Yuan et al. 2018), low concentrations of cations and a low cation exchangeable capacity (CEC) (Asensio et al. 2019; Zhen et al. 2019). Total organic carbon contents in the range of 5.6 15.9 g kg^{-1} (Kumar et al. 2018) and the high bioavailability of metalloid elements such as Cd, Pb, Cu, Ni, and Zn (Manna and Maiti 2018; Pietrzykowski et al. 2014) are observed in RMSs. Therefore, soil amelioration is essentially required to improve the quality of RMSs before revegetation is conducted in the soils.

In general, the application of ash to soils as a form of soil amelioration improves the soil physical, chemical, and biological characteristics (Moragues-Saitua et al. 2017; Thomaz 2018). The application of 3–6 Mg ha^{-1} wood ash to soils increased the pH and nutrient concentrations in the O horizon of forest soil after 2.5 years (Hansen et al. 2018). Furthermore, the application of whole digestate combined with wood ash to soils resulted in higher soil pH and nitrate concentration compared to the application of whole digestate without wood ash (Ibeto et al. 2020). However, the results of a study carried out by García-Sánchez et al. (2015) showed that no significant changes in soil chemical and microbiological parameters followed coal fly ash application to Chernozem soil. No changes in C mineralization and urease activity in soils were observed after 14 years of coal fly ash application (Leclercq-Dransart et al. 2019). Differences in the effect of ash used as an amelioration material on changes in soil properties may be attributed to differences in the type and amount of elements contained in the ash. Until now, comprehensive information on the changes in soil properties resulting from different ash applications to RMSs has been relatively unavailable. The lack of such information restricts the practical use of ash as an amelioration material to improve the characteristics of RMSs. The aim of this study was to evaluate the changes in C mineralization and MBC in response to different ash applications to RMSs. The hypothesis is that the application of different ash to RMSs results in different C mineralization and soil MBC values.

2 Materials and Methods

2.1 Study Site Description

The research site was in the reclaimed coal mining area at the PT Arutmin Indonesia Satui site (03°11′27″ – 03°46′41″ S, 115°22′56″ – 115°54′14″ E) in the South Kalimantan Province, Indonesia. Soils at this site are classified as Typic Dystrudepts on the basis of the soil taxonomy system. The average annual precipitation is 3001 mm, ranging from 1157 to 4459 mm, with approximately 75% precipitation between March and July. The average annual temperature is 27.5 °C, with a mean minimum temperature of 22.7 °C and a mean maximum temperature of 31.9 °C observed in June and November, respectively.

The research site is a former coal mine area with an open pit system that was closed in 2009. After the closure of the mine, the site was reclaimed and planted with forestry plants and cover crops. The dominant trees at the site include *Acacia mangium* and *Paraserianthes falcataria*. Smaller proportions of *Samanea saman*, *Shorea megistophylla*, *Dipterocarpus hasseltii,* and *Peronema canescens* were observed at the site. The cover crop at this site is *Calopogonium mucunoides*.

2.2 Soil and Ash Sampling and Characterization

The soil used for this study was characterized by the O horizon (4–0 cm), A horizon (0–45 cm) and B horizon (45–90 cm). The organic layer (O horizon) is dominated by slightly to moderately decomposed litter originating from *C. mucunoides,* which is used as a cover crop in the reclamation of coal mine soils. Soil samples were collected randomly from 20 soil cores at a depth of 0–30 cm (A horizon) using a soil auger, and then the samples were combined into a soil composite sample. Plant debris was removed manually, and the soil samples were homogenized, placed in a plastic bag, and stored under field-moist conditions at 4 °C until they were used for the incubation study. Soil subsamples were air-dried at room temperature and sieved to 2 mm for soil physical and chemical characterization.

Rice husk ash was sampled from rice mills located in the Asam-Asam Village, Jorong Subdistrict, South Kalimantan Province, which is adjacent to the reclaimed coal mining area of the PT Arutmin Indonesia Satui site. Oil palm shell ash was collected from the PTPN XIII oil palm processing plant, Pelaihari, South Kalimantan Province, Indonesia. Coal fly ash was sampled from the Asam-Asam Coal Power Plant, which is located in Asam-Asam village adjacent to the reclaimed coal mining area of the PT Arutmin Indonesia Satui site. All ashes were air-dried after collection and then homogenized by sieving (2 mm mesh) before physical and chemical characterization.

2.3 Organic Matter Preparation

Calopogonium mucunoides grown in the reclaimed coal mining area of the PT Arutmin Indonesia Satui site as a cover crop was used as OM in this study. Plant residues were oven-dried at 60 °C and then ground to <2 mm. The residues contained 370.4 g kg^{-1} organic carbon, 28.5 g kg^{-1} total nitrogen, 27.3 g kg^{-1} hot water-soluble carbon, 37.2 g kg^{-1} cellulose, 31.5 g kg^{-1} hemicellulose, and 14.9 g kg^{-1} lignin (Saidy et al. 2019).

2.4 Laboratory Incubation

The experiment was conducted using polyvinyl chloride (PVC) tubes (1.95 cm diameter) containing 30.40 g of moist soil (the amount of soil in the tube was calculated to obtain a bulk density of soil similar to that in field measurements after the compaction of the soil to a depth of 2.00 cm). OM (0.3 g) and ash (0.00, 0.36, 0.72, and 1.07 g) were added gently to and combined homogenously with the soil. The amount of OM added to the soil was equivalent to a field application of 5.0 Mg ha^{-1}, while the amount of ash added to the soil was equivalent to field applications of 0, 25, 50, and 75 Mg ha^{-1}, respectively. Distilled water was added carefully to the mixture of soil OM and ash to achieve 70% water-filled pore space (WFPS), and the mixture in the tube was compacted to a depth of 2.00 cm. The tubes were then transferred to 1000 mL Mason jars along with 15 mL of distilled water in a 20 mL glass vial for humidity maintenance. After being sealed with airtight lids with rubber septa for gas sampling, the jars were incubated in the dark at room temperature for 120 days. A total of 36 tubes were prepared and incubated: three ashes×four levels of ash application×three replicates.

Carbon mineralization was determined by extracting 10 mL of headspace gas from each jar using a 10 mL syringe through the septum in the middle of the lid. The extracted gas was transferred to a 10 mL glass vial and then injected onto a gas chromatograph (Shimadzu GC-14A). Carbon mineralization was measured on a weekly basis during the 120-day incubation period. After the C mineralization measurement was completed each week, the jars were opened for 3 h to permit the exchange of the CO_2-enriched water inside the jar with fresh water. The tubes were watered precisely when the jars were opened to ensure a constant water content during the incubation period (the water content at the end of incubation was 69.2–71.2% WFPS). The total C mineralization in each sample during the incubation period of 120 days was calculated as the sum of each C mineralization measurement every week and expressed as the cumulative C mineralization.

2.5 Characterization of Soil, Ash, and Amended Soil

The determination of soil texture was conducted using sieving and sedimentation methods (Gee and Bander 1986). The bulk density of the soil and ash was determined by driving a cylindrical metal sampler (diameter = 4.8 cm; height = 10.0 cm) into the soil or ash to a 30 cm depth, and then the cylindrical metal was carefully removed to preserve the soil and ash cores (Blake and Hartge 1986). The soil and ash samples were transported to the laboratory, dried to a constant weight at 105 °C and then weighed. The soil and ash pH values were determined using the electrode glass method in an aqueous mixture of air-dried sample and distilled water (1:5, mass:volume) (McLean 1982). The contents of organic carbon and total nitrogen in the soil and the ash were measured by the dry combustion method using a LECO CNS2000 (LECO Corporation, MI, USA). The total P of the soil and ash was measured using molybdenum blue with spectrophotometric measurements at 660 nm after digestion of the soil and ash with 60% $HClO_4$ (Olsen and Sommers 1982). CEC was measured using the ammonium acetate (pH 7.0) method (Rhoades 1982). Measurements of total potassium (K), sodium (Na), magnesium (Mg), calcium (Ca), aluminum (Al), and iron (Fe) were conducted by digestion of the soil and ash using a mixture of HNO_3 and $HClO_4$ in glass test tubes for 2 h at 100 °C, followed by digestion at 120 °C until a white residue was obtained. The solution containing the white residue was diluted with distilled water to 50 mL and then filtered through Whatman No. 41 filter paper (Barnhisel and Bertsch 1982; Olson and Ellis 1982). The concentrations of K, Na, Ca, Mg, Al, and Fe in the solution were determined using atomic absorption spectrophotometry (Shimadzu AA6300G). The characteristics of the soil and ash samples in the study are presented in Table 1.

Selected soil chemical properties were determined following the completion of the incubation period. The soil reaction (pH), CEC, and total Ca and Mg of the ash-amended soils were measured using the methods described previously. The specific surface areas (SSA) of the soils were acquired by five-point nitrogen adsorption at 77 K and the subsequent desorption of nitrogen with an autosorb instrument (Nova 4200 Analyzer, Quantachrome Corp., Boynton Beach, USA). The extraction of Fe and Al from the soils using oxalate (Fe_o and Al_o) and dithionite (Fe_d) solutions was conducted by the method of Blakemore et al. (1987), and the concentrations of Fe and Al in the extracts were quantified spectrophotometrically (Shimadzu AA6300G).

After the completion of the incubation period, all tubes were removed from the jars, and the ash-amended soils were air-dried for 6 h to determine the soil MBC. Soil MBC was measured by the chloroform fumigation-extraction (CFE) method (Vance et al. 1987), and a KEC value of 0.45 was used to measure MBC (Joergensen 1996). Two soil subsamples of 5 g each (approximately 40% water holding capacity), one designated for nonfumigation and the other for fumigation, were placed into a 50 mL conical flask. The soil samples were fumigated using ethanol-free chloroform in a vacuum desiccator for 24 h at room temperature in the dark. Both the nonfumigation and fumigation samples were extracted by adding 40 ml of 0.5 M K_2SO_4 to the soil and were shaken on a shaker at 40 cycles per minute for 30 min. The extract was filtered through Whatman No. 41 filter paper, and the

Table 1 Characteristics of the soil and different types of ash used in the experiment

Characteristics	Soil	*Coal fly ash*	Rice husk ash	Oil palm shell ash
Texture				
Sand (%)	38.33 (3.99) *	–	–	–
Silt (%)	23.45 (3.72)	–	–	–
Clay (%)	38.22 (2.56)	–	–	–
pH (H_2O)	4.23 (0.12)	7.12 (0.10)	6.45 (0.09)	6.68 (0.09)
Bulk density (g cm^{-3})	1.21 (0.03)	1.37 (0.10)	1.27 (0.10)	1.19 (0.04)
Organic C (g kg^{-1})	2.34 (0.13)	1.02 (0.05)	1.12 (0.07)	1.24 (0.06)
N (g kg^{-1})	1.45 (0.03)	0.91 (0.04)	0.34 (0.04)	0.19 (0.03)
P (g kg^{-1})	1.67 (0.09)	0.11 (0.03)	0.09 (0.04)	0.07 (0.02)
Ca (mg kg^{-1})	2.34 (0.13)	1897.30 (8.73)	23.56 (2.83)	45.23 (6.24)
Mg (mg kg^{-1})	3.23 (0.05)	1684.30 (7.08)	43.44 (5.50)	36.22 (2.04)
K (mg kg^{-1})	2.12 (0.06)	678.23 (6.10)	3.45 (0.15)	3.12 (0.04)
Na (mg kg^{-1})	1.23 (0.08)	512.30 (5.42)	1.23 (0.04)	1.57 (0.09)
Al (mg kg^{-1})	12.34 (0.10)	823.21 (4.65)	3.44 (0.16)	3.13 (0.08)
Fe (mg kg^{-1})	9.67 (0.51)	532.31 (2.98)	2.56 (0.11)	2.34 (0.06)
CEC (cmol kg^{-1})	19.87 (0.08)	–	–	–

*Numbers in the parentheses indicated the standard deviation of mean ($n = 3$)

contents of organic C in the extract were measured using the Walkley-Black wet digestion method (Heanes 1984). Soil MBC was calculated by subtracting the nonfumigated C measurement from the fumigated C measurement.

2.6 Carbon Mineralization Fitting and Statistical Analysis

The C mineralization data were fitted to the two-pool carbon mineralization model, i.e., $C_t = C_s\,(1\text{-}e^{-st}) + C_f\,(1\text{-}e^{-ft})$, to quantify the dynamics of C mineralization, where C_t is the cumulative C mineralization (mg C kg^{-1}) during the incubation period t; C_s is the size of the pool of slowly mineralizable C (mg C kg^{-1}); C_f is the size of the pool of rapidly mineralizable C (mg C kg^{-1}); and s and f are the mineralization rate constants for the slow and fast pools (day^{-1}), respectively. Curve fitting was carried out by the least-squares nonlinear curve fitting procedure in Microsoft Excel® (de Levie 2001).

The experimental data were analyzed by analysis of variance (ANOVA) in GenStat 11th Edition to test the effects of each treatment (Payne 2008). In the case of significance in ANOVA, the least significant difference (LSD) test was used to differentiate among the treatment means at the 95% confidence level.

3 Results

3.1 Characteristics of Soils with Different Types and Amounts of Added Ash

The ANOVA results revealed that the addition of ash to the RMSs resulted in significant changes in several soil chemical characteristics, except soil CEC (Table 2). The increase in soil pH was larger with coal fly ash addition than with oil palm shell and rice husk ash addition ($P \leq 0.05$) (Table 3), suggesting that the neutralizing values of coal fly ash were higher than those of both oil palm shell and rice husk ash. The SSA of the soils increased with ash addition. The largest increase in SSA was observed for coal fly ash addition (Table 4). The contents of Ca, Mg, oxalate-extractable Al, oxalate-extractable Fe and dithionite-extractable Fe in the soil also increased with the addition of ash, with higher increases after coal fly ash addition than after oil palm shell and rice husk ash addition (Table 4).

3.2 Effect of Ash Additions on Carbon Mineralization

Rapid C mineralization was observed for all treatments over the first 35 days; then, the C mineralization increased gradually, starting to flatten on day 98 (Fig. 1A, B, and C). The cumulative carbon mineralization at the end of the incubation period reached 1260–1593 mg C kg^{-1}, depending on the type and amount of ash added to the soil (Fig. 1D).

The ANOVA results showed that the cumulative C mineralization during the 120-day incubation period was significantly influenced by ash application ($P \leq 0.001$; Table 2). The C mineralization of soils amended with oil palm shell and rice husk ash exhibited similar responses to increasing amounts of applied ash. Increasing the amount of oil palm shell or rice husk ash to high rates (50 and 75 Mg ha^{-1}) increased C mineralization ($P \leq 0.05$; Fig. 1D). However, the response of C mineralization to increasing amounts of added coal fly ash was different from that to increasing amounts of added oil palm shell and rice husk ash.

Table 2 Results of nested ANOVA of the effect of different types and amounts of ash addition on changes in soil characteristics, C mineralization, parameters of the two-pool C mineralization model (C_s, C_f, s, and f), and MBC

Source of Variation	Degrees of Freedom	F	*P* value
Soil pH			
Ash type	2	19.69	<0.001
*Ash type*Ash amount*	9	22.80	<0.001
CEC			
Ash type	2	19.69	0.201
*Ash type*Ash amount*	9	22.80	0.134
Ca			
Ash type	2	224.62	<0.001
*Ash type*Ash amount*	9	101.17	<0.001
Mg			
Ash type	2	170.69	<0.001
*Ash type*Ash amount*	9	40.28	<0.001
SSA			
Ash type	2	57.29	<0.001
*Ash type*Ash amount*	9	22.57	<0.001
Al_O			
Ash type	2	155.14	<0.001
*Ash type*Ash amount*	9	70.77	<0.001
Fe_O			
Ash type	2	199.28	<0.001
*Ash type*Ash amount*	9	44.34	<0.001
Fe_D			
Ash type	2	229.67	<0.001
*Ash type*Ash amount*	9	81.11	<0.001
C mineralization			
Ash type	2	31.50	<0.001
*Ash type*Ash amount*	9	9.55	<0.001
C_f			
Ash type	2	36.55	<0.001
*Ash type*Ash amount*	9	10.57	<0.001
C_s			
Ash type	2	23.48	<0.001
*Ash type*Ash amount*	9	11.92	<0.001
f			
Ash type	2	47,286.45	<0.001
*Ash type*Ash amount*	9	17,973.82	<0.001
s			
Ash type	2	147,000.00	<0.001
*Ash type*Ash amount*	9	72,180.00	<0.001
Microbial biomass C			
Ash type	2	6.16	0.007
*Ash type*Ash amount*	9	5.62	<0.001

The application of 75 Mg ha^{-1} coal fly ash led to a decrease of 14% in C mineralization compared to the soil without coal fly ash application ($P \leq 0.05$; Fig. 1D).

The cumulative C mineralization of soil amended with ash fitted very well to the two-pool carbon mineralization model with $R^2 \geq 0.99$ (Fig. 1A, B and C). The size of the rapidly mineralizable pool (C_f − 855-1102 mg C kg^{-1}) was larger than that of the slowly mineralizable pool (C_s – 429-578 mg C kg^{-1}) (Table 5). The addition of oil palm shell and rice husk ash to the RMSs resulted in increases in both C_f and C_s, but the increase in C_f was stronger than that in C_s (Table 5). The greatest increase in the size of C_f by 11% and 15% was observed when the amount of added oil palm shell and rice husk ash reached 75 Mg ha^{-1}, respectively. The addition of 50 Mg ha^{-1} oil palm shell and rice husk ash resulted in the greatest increase in the size of C_s by 8% and 12%, respectively (Table 5). However, the addition of coal fly ash to the RMSs decreased both C_s and C_f (Table 5). The results of ANOVA revealed that the mineralization rates of the slow (s) and fast (f) mineralizable pools were not significantly influenced by the addition of different types and amounts of ash ($P > 0.05$; Table 2).

3.3 Microbial Biomass Carbon

The ANOVA results showed that the soil MBC was influenced by the application of ash ($P \leq 0.001$). The addition of oil palm shell and rice husk ash at 50 Mg ha^{-1} and 75 Mg ha^{-1} to the RMSs increased MBC compared to that of soil without ash ($P \leq 0.05$; Fig. 2). In contrast, the addition of a high amount of coal fly ash to the RMSs led to a reduction in MBC. The addition of 25 Mg ha^{-1} coal fly ash increased MBC. However, the MBC of the soil with 75 Mg ha^{-1} coal fly ash was lower than that of the soil without ash ($P \leq 0.05$; Fig. 2).

4 Discussion

The application of different types and amounts of ash to the RMSs resulted in similar effects on C mineralization and MBC. The application of different amounts of rice husk and oil palm shell ash and a low amount of coal fly ash increased C mineralization and MBC, while the application of a high amount of coal fly ash reduced C mineralization and MBC. The results of this study are consistent with a study by Basanta et al. (2017) that reported changes in both microbial activity and microbial biomass after the application of remediation treatments. Ash application to soils indirectly influences soil C mineralization and MBC through changes in the activity of soil microorganisms. Increases in soil pH as a result of ash application have been reported to accelerate microbial activity and therefore increase C mineralization and soil MBC (Barthod et al. 2018; Cruz-Paredes et al. 2017; Maljanen et al. 2006; Reid and Watmough 2014; Zimmermann and Frey 2002). Such an increase in soil MBC due to changes in

Table 3 Changes in pH, cation exchangeable capacity (CEC), calcium (Ca), and magnesium (Mg) of soils after the addition of different types and amounts of ash

Type of ash	Amount	pH (H_2O)	CEC	Ca	Mg
	(Mg ha^{-1})		(cmol kg^{-1})	— g kg^{-1} —	
Oil palm shell ash	0	3.63 (0.09)* a**	21.87 (0.97)	0.77 (0.11) a	1.11 (0.13) a
	25	3.68 (0.10) ab	20.54 (1.57)	1.10 (0.12) bc	1.23 (0.12) a
	50	3.94 (0.05) cd	20.87 (2.37)	1.63 (0.17) d	1.61 (0.15) b
	75	4.06 (0.07) d	20.23 (1.86)	1.99 (0.13) e	1.96 (0.17) c
Rice husk ash	0	3.64 (0.06) a	20.98 (3.16)	0.77 (0.11) a	1.11 (0.13) a
	25	3.71 (0.16) ab	19.54 (1.75)	0.99 (0.13) ab	1.25 (0.24) a
	50	3.91 (0.06) cd	19.87 (1.43)	1.34 (0.11) c	1.26 (0.15) a
	75	4.06 (0.08) d	18.76 (1.64)	1.83 (0.15) de	1.31 (0.06) a
Coal fly ash	0	3.64 (0.07) a	22.04 (1.99)	0.77 (0.11) a	1.11 (0.13) a
	25	3.84 (0.05) bc	20.54 (2.57)	1.92 (0.24) e	2.26 (0.25) d
	50	4.23 (0.12) e	18.65 (1.98)	3.26 (0.30) f	2.96 (0.17) e
	75	4.48 (0.18) f	16.45 (2.15)	4.15 (0.19) g	3.27 (0.17) f

*Numbers in the parenthesis represent the standard deviation of mean (n = 3)

**Lower cases following the standard deviation indicate no significant differences among the treatments based on the LSD test at $P \leq 0.05$

soil pH is primarily related to the increase in soil bacterial activity (Rousk et al. 2010). In this experiment, all ash additions increased the soil pH from 3.60 to 4.06–4.48, depending on the types and the amounts of added ash ($P \leq 0.05$; Table 3), indicating that the ash used in this experiment could be an alternative material for liming acid soils (Pandey and Singh 2010; Schönegger et al. 2018).

It is known that MBC shows a quick response to soil amelioration and plays an essential function in controlling changes in soil quality (Bünemann et al. 2018; Kiboi et al. 2018). The increase in MBC in response to the application of different amounts of rice husk and oil palm shell ash and a low amount of coal fly ash in this study might be because of the easily available nutrients in the ash. The ash used in this experiment contains nutrients (Table 1) necessary for the growth of soil microorganisms, although not all of the nutrients are present in an available form. The results of this research are in line with a study conducted by Jokinen et al. (2006) that showed increasing amounts and qualities of available C sources (dissolved organic C) for microbial activity following the application of wood ash. The amount of C in microbial biomass and the amount of K_2SO_4-extractable dissolved organic C was higher in soil treated with wood ash and nitrogen applications than in soil treated with nitrogen application only, indicating that wood ash application resulted in an increase in the amount of available C for microorganisms (Saarsalmi et al. 2012).

Table 4 Changes in specific surface areas (SSA), oxalate-extractable aluminum (Al_O), oxalate-extractable iron (Fe_O), and dithionite-extractable iron (Fe_D) of soils after the addition of different types and amounts of ash

Type of ash	Amount	SSA	Al_O*	Fe_O*	Fe_D**
	(Mg ha^{-1})	(m^2 g^{-1})	— g kg^{-1} —		
Oil palm shell ash	0	11.07 (1.09)* a**	1.11 (0.13) a	1.16 (0.19) a	1.57 (0.10) a
	25	12.28 (0.25) ab	1.46 (0.11) bc	1.19 (0.02) a	2.11 (0.13) b
	50	13.30 (0.28) bc	1.88 (0.10) d	1.33 (0.12) ab	2.30 (0.17) bc
	75	13.50 (0.95) cd	2.12 (0.13) e	1.45 (0.11) abc	2.55 (0.10) c
Rice husk ash	0	11.07 (1.09) a	1.11 (0.13) a	1.16 (0.19) a	1.57 (0.10) a
	25	11.98 (0.65) ab	1.27 (0.08) ab	1.31 (0.17) ab	1.99 (0.13) b
	50	12.22 (0.24) bc	1.56 (0.12) c	1.59 (0.14) bc	2.13 (0.19) b
	75	12.34 (0.85) cd	2.00 (0.13) de	1.66 (0.28) c	2.14 (0.17) b
Coal fly ash	0	11.07 (1.09) a	1.11 (0.13) a	1.16 (0.19) a	1.57 (0.10) a
	25	15.48 (1.10) d	2.18 (0.24) e	2.30 (0.17) d	2.99 (0.13) d
	50	18.11 (0.50) e	3.02 (0.17) f	3.12 (014) e	3.88 (0.39) e
	75	19.70 (1.55) f	3.30 (0.13) g	3.85 (0.28) f	5.52 (0.33) f

*Numbers in the parenthesis represent the standard deviation of mean ($n = 3$)

**Lower cases following the standard deviation indicate no significant differences among the treatments based on the LSD test at $P \leq 0.05$

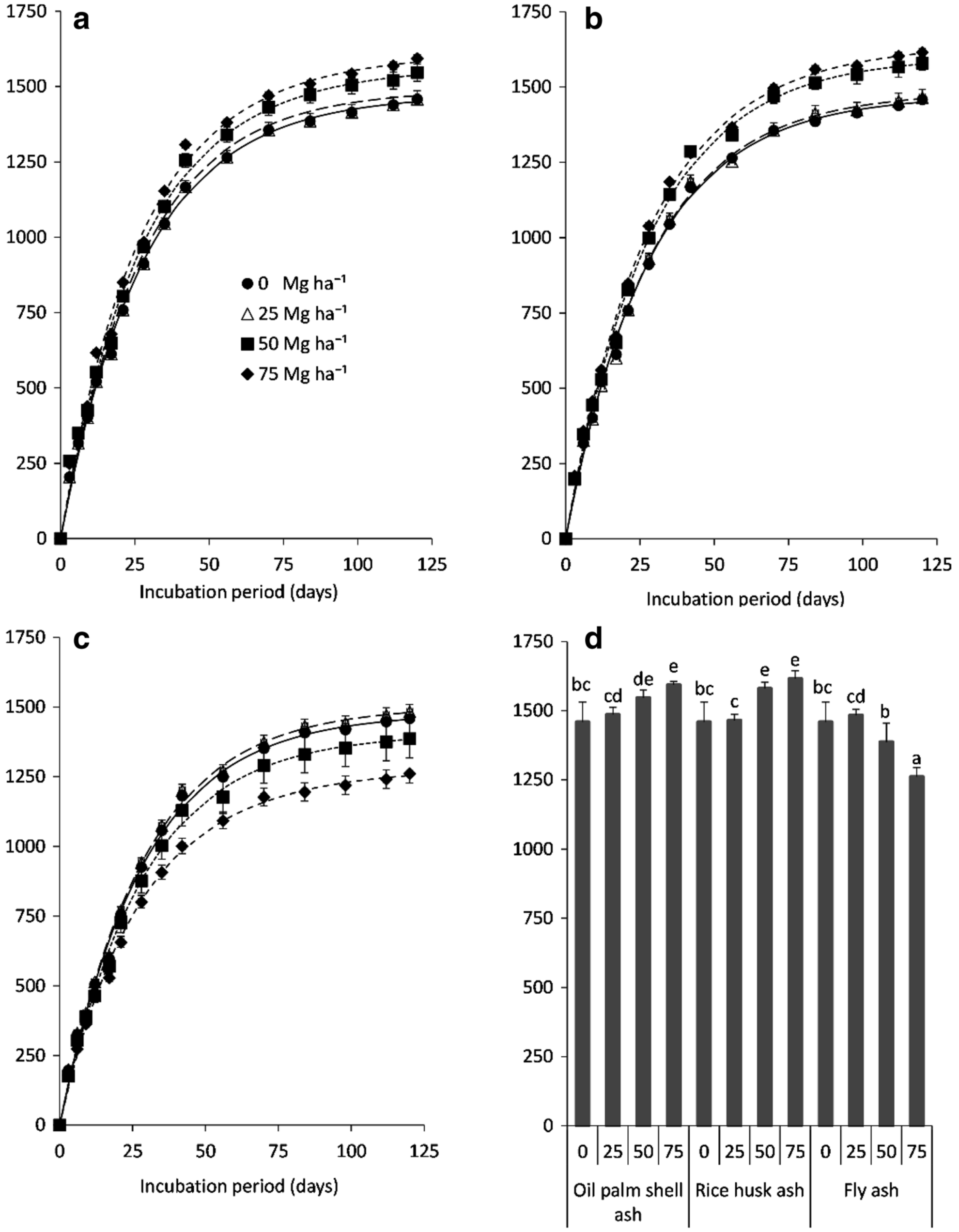

Fig. 1 Carbon mineralization of reclaimed-mine soil with different amounts of oil palm shell ash (A), rice husk ash (B), and coal fly ash (C), throughout a 120-day incubation period. Vertical bars represent the standard deviation of the mean ($n = 3$). The lines are curves fitted to the two-pool carbon mineralization model: $C_t = C_s(1-e^{-st}) + C_f(1-e^{-ft})$; $R^2 > 0.98$. Cumulative C mineralization of reclaimed-mine soil with additions of different ash throughout a 120-day incubation period (D). Different lowercase letters above the columns indicate significant differences between the treatments (LSD test, $P < 0.05$)

Increased inorganic nitrogen has been observed following wood ash application to soils (Vestergård et al. 2018), suggesting increased amounts of easily available nutrients for soil microorganisms. This result indicates that the application of rice husk and oil palm shell ash and a low amount of fly ash to RMSs increases soil biochemical processes that eventually improve the characteristics of the RMSs.

The effect of coal fly ash addition on C mineralization and soil MBC is different from that of rice husk and oil palm shell ash addition on the soil MBC; i.e., an increase in soil MBC occurred at the lowest amount of added coal fly ash, while C mineralization and soil MBC decreased at higher amounts of added coal fly ash. The changes in the effects of low and high amounts of coal fly ash on C mineralization and MBC are attributed to the direct effect of coal fly ash on easily available nutrients for microorganisms (Nayak et al. 2014). An increase in C mineralization and soil MBC at low amounts of added coal fly ash may be related to the presence of low amounts of Fe, Al, and other elements derived from coal fly ash that function as easily available nutrients in soil biochemistry. However, the decrease in MBC under high amounts of added coal fly ash may be attributed to the detrimental effect of the metals contained in coal fly ash. Coal fly ash contains relatively high amounts of Fe and Al (Table 1), which may hinder soil microbial activity. The low C mineralization of bentonite waste is attributed to the presence of a high concentration of

Table 5 Results of carbon mineralization data fit to the two-pool mineralization model

Type of ash	Amount of ash (Mg ha^{-1})	C_s (mg C kg^{-1})	s (day^{-1})	C_f (mg C kg^{-1})	f (day^{-1})	R^2
Oil palm shell ash	0	510.44 (25.37)* b**	0.0308 (0.00075)	963.18 (47.73) b	0.0378 (0.00002)	0.999
	25	508.25 (14.78) b	0.0234 (0.00008)	1005.77 (13.84) bc	0.0430 (0.00014)	0.999
	50	550.25 (10.25) de	0.0276 (0.00012)	1015.62 (18.92) c	0.0402 (0.00005)	0.999
	75	545.00 (4.43) cd	0.0248 (0.00009)	1070.35 (8.69) de	0.0431 (0.00008)	0.999
Rice husk ash	0	517.62 (25.81) bc	0.0350 (0.00034)	960.23 (47.60) b	0.0350 (0.00007)	0.999
	25	519.72 (7.63) bc	0.0350 (0.00007)	964.33 (14.17) b	0.0350 (0.00002)	0.999
	50	577.84 (2.54) e	0.0350 (0.00018)	1022.83 (4.50) cd	0.0350 (0.00021)	0.999
	75	542.78 (9.72) cd	0.0258 (0.00005)	1102.44 (19.74) e	0.0403 (0.00012)	0.999
Coal fly ash	0	517.62 (25.54) bc	0.0350 (0.00008)	960.23 (47.60) b	0.0350 (0.00008)	0.999
	25	526.11 (7.81) bcd	0.0350 (0.00015)	976.19 (14.49) bc	0.0350 (0.00014)	0.999
	50	507.25 (25.01) bc	0.0350 (0.00008)	897.47 (44.61) a	0.0350 (0.00008)	0.999
	75	429.37 (10.70) a	0.0234 (0.00011)	855.40 (23.37) a	0.0430 (0.00016)	0.999

*Numbers in the parenthesis represent the standard deviation of mean (n = 3)

**Lower cases following the standard deviation indicate no significant differences among the treatments based on the LSD test at $P \leq 0.05$

toxic compounds in the bentonite waste (Rodríguez-Salgado et al. 2017). A large body of research in the last decade has been carried out to elucidate the changes in soil properties due to coal fly ash application (Pandey and Singh 2010). Briefly, studies on the effect of a low amount of coal fly ash on soil MBC have shown conflicting results. Schönegger et al. (2018) reported that the addition of a low amount of coal fly ash (2 *w*/w %) to soils resulted in the suppression of soil MBC after 60 and 100 days of incubation. The soil MBC and soil enzymatic activity did not change following a low amount of coal fly ash application (15 Mg coal fly ash ha^{-1}) to soils (García-Sánchez et al. 2015). In another study, Lim and Choi (2014) found that the soil MBC observed 7 days after incubation increased significantly following the application of a low amount of coal fly ash (5–10 *w*/w %) to the soil.

In contrast, no conflicting results on the reduction in soil MBC following the application of a relatively high amount of coal fly ash have been reported in previous studies (e.g., Nayak et al. 2014; Parab et al. 2015). Parab et al. (2015) reported that the number of soil beneficial microbes (*Azotobacter*) declined considerably with coal fly ash application, while the soil microbial activity substantially increased with the application of up to 50 Mg ha^{-1} coal fly ash and then decreased when the amount of applied coal fly ash reached 100 Mg ha^{-1}. A study conducted by Parab et al. (2015) revealed that 50 Mg ha^{-1} is considered an optimal amount of applied coal fly ash for improving soil microbial properties. In another study, Nayak et al. (2014) found that the soil MBC decreased significantly throughout a 120-day incubation period with 10% 20% coal fly ash addition to soils, which might be caused by a reduction in substrate availability due to the accumulation of persistent lignite-derived organic carbon compounds. Decreases in MBC may also be related to the decrease in the soil microbial population with the application of high amounts of coal fly ash to soils (Pandey and Singh 2010). Soluble C and enzyme activities (alkali phosphatase, arylsulfatase, b-glucosidase, and L-asparaginase) decrease significantly in co-composted public green waste with high coal fly ash application rates (Belyaeva and Haynes 2009).

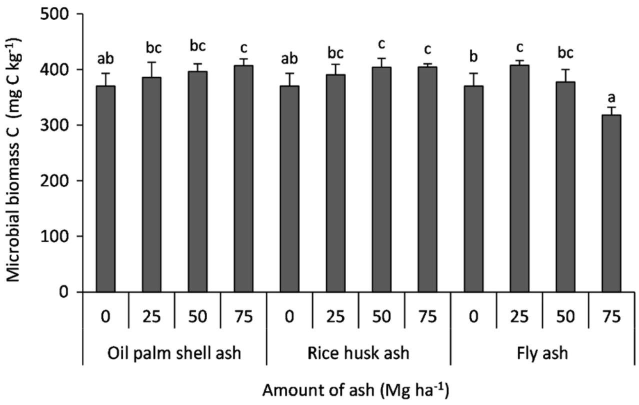

Fig. 2 Microbial biomass C of reclaimed-mine soil with the addition of different types of ash. Vertical bars indicate the standard deviation of the mean ($n = 3$). Different lowercase letters above the columns indicate significant differences between the treatments (LSD test, $P < 0.05$)

Another possible mechanism for the decrease in the C mineralization of soils with high amounts of coal fly ash is the reduction in the availability of organic carbon for microorganisms through soil physicochemical reactions, i.e., the stabilization of organic C by oxides contained in coal fly ash. The application of a high amount of coal fly ash to soil resulted in increases in the contents of Fe_O, Al_O, and Fe_D (Table 4). It is well known that the presence of Fe_O, Al_O, and Fe_D (iron and aluminum oxides) in soils increases the sites (specific surface areas) for OC sorption supplied by the high density of reactive surface functional groups associated with those oxides. It has been suggested that the higher SSA of soil with coal fly ash than that of control soil (Table 2) may enable more organic carbon-soil interactions and thereby result in a reduction in carbon mineralization (Saidy et al. 2012; von Lützow et al. 2006; Wattel-Koekkoek et al. 2003). Decreases in C mineralization with high levels of coal fly ash addition have also been found in previous experiments (McCarty et al. 1994; Nayak et al. 2014; Pandey and Singh 2010; Pitchel 1990). Lim et al. (2012) suggested that the application of coal fly ash can reduce C emissions due to the formation of carbonate from CO_2 resulting from the C mineralization facilitated by calcium-enriched coal fly ash.

The reduction in the C mineralization of the RMSs with high amounts of coal fly ash is also related to the presence of polyvalent cations in coal fly ash. Coal fly ash has high contents of calcium and magnesium (Table 1), which increase the sorption of OM through the mechanism of cation bridging. The sorption of OM increases the amount of OM protected from soil microbial decomposition. The calcium and magnesium cations from the added coal fly ash, which increase considerably with increasing coal fly ash application (Table 3), may function as bridges between the negatively charged functional groups of OM and negatively charged clay minerals, ultimately leading to increase OM sorption (Arnarson and Keil 2000; Feng et al. 2005; Singh et al. 2016).

Changes in the C mineralization of the RMSs due to ash application may also be attributed to alterations in the size of the slowly mineralizable pool, rapidly mineralized pool or both pools. The application of oil palm shell and rice straw ash increased C mineralization, and the sizes of both the slowly and rapidly mineralizable pools increased substantially with ash application. In contrast, the sizes of both the slowly and rapidly mineralizable pools decreased with coal fly ash application, and C mineralization decreased with coal fly ash application. These observations suggest that coal fly ash application results in the protection of a relatively large proportion of OM.

Ash application resulted in either increased C mineralization or decreased C mineralization, and the mineralization rates of both the slowly and rapidly mineralized pools did not change with ash application. This result indicates that ash addition affected the dynamics of C mineralization by changing the size of mineralizable C pools. This result is in line with the previous finding that C mineralization was reduced significantly for clays coated with iron and aluminum oxides; the sizes of the slowly and rapidly mineralizable pools were considerably reduced, while the mineralization rates of these pools were unaffected (Saidy et al. 2012). Previous studies have shown that the size of the mineralizable C pool, obtained from the two-pool C mineralization model, increases significantly after the sorption of organic C onto soils (Kalbitz et al. 2005), clay minerals and goethite (Mikutta et al. 2007).

The results of this study showed that the addition of oil palm shell and rice husk ash at all doses and coal fly ash at a low dose resulted in increases in C mineralization and soil MBC, while C mineralization and soil MBC decreased with a high amount of coal fly ash application to the RMSs. Therefore, our hypothesis that differences in the characteristics of the added ash lead to different C mineralization and MBC in the RMSs was supported. The different effects of ash application on C mineralization and soil MBC also imply that coal fly ash application to the RMSs at high amounts may protect soil C from microbial decomposition and thereby increase soil C stabilization. With regard to ash management, the results of this study indicate that ash could be applied as a waste material to the RMSs to improve soil properties and that the extent of the effect of ash application on changes in soil properties varied with the ash characteristics.

5 Conclusions

The results of the study showed that the application of different types of ash to the reclaimed-mining soils resulted in different carbon mineralization and soil microbial biomass carbon effects. The effect of oil palm shell and rice straw ash application on carbon mineralization was different from that of coal fly ash application. The addition of oil palm shell and rice straw ash to the reclaimed-mining soils increased carbon mineralization. Low amounts of coal fly ash application to the soils enhanced carbon mineralization, but C mineralization decreased with a high amount of coal fly ash application. This decrease in C mineralization may be attributed to increases in the contents of Ca, Mg, Fe_o, and Al_o (oxalate-extractable iron and aluminum), and Fe_d (dithionite-extractable iron), as a high amount of applied coal fly ash may increase the stabilization of soil organic matter. The increased specific surface areas (SSA) for organic carbon sorption provided by surface functional groups associated with oxides contained in the coal fly ash may also lead to a decrease in C mineralization. The results of the carbon mineralization data fitted to the two-pool carbon mineralization model showed that changes in soil carbon mineralization in response to ash application are associated with differences in the sizes of the mineralizable carbon pools (rapidly mineralizable carbon pool, slowly mineralizable carbon pool or both pools). In the oil palm shell and rice straw ash treatments that increased C mineralization, the mineralizable C pool size increased with the application of oil palm shell and rice straw ash to the soil. However, the application of coal fly ash to soils led to a decrease in the size of the mineralizable C pool, which eventually resulted in decreases in C mineralization. This result demonstrates that coal fly ash application to the reclaimed-mining soils leads to the protection of a relatively large proportion of organic matter, thereby increasing the organic matter content in the reclaimed mining soils. Similar to carbon mineralization, the soil microbial biomass carbon of the reclaimed mining soils increased significantly with oil palm shell and rice husk ash application. However, the addition of a high amount of coal fly ash (75 Mg ha^{-1}) led to a reduction in the soil microbial biomass carbon compared to that of soils without coal fly ash application. With regard to ash management, the results of this study indicate that the application of ash to improve the soil properties of the reclaimed mining soils could be used as an alternative to managing ash as a waste material.

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Compliance with Ethical Standards

Conflict of Interest The authors declare that they have no conflict of interest.

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