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Selection of organic matter as a wetland substrate for acid mine drainage treatment

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Abstract*. Acid mine drainage (AMD) is one of the problems arising from mining activities. Acid mine drainage is formed due to the oxidation of sulfide minerals such as pyrite (FeS_2) by water and oxygen. An alternative treatment of AMD in a sustainable and eco-friendly way is by constructing a wetland. This study was conducted to select the best composition of organic matter capable of treating acid mine drainage to comply with environmental quality standards. The study consisted of three stages, namely; a screening of organic matter, a combination of two organic matters, and a combination of cow manure and empty fruit bunches (EFB) in various compositions. Types of organic matter used are cow, goat, and chicken manure. Three types of compost, EFB, sawdust, wood chips, chopped water hyacinth, cocopeat, fresh waste, and compost waste cajuputi leaf, waste of citronella distillation, baglog waste, and bagasse. The results showed that several types of organic matter could increase the pH of AMD. The combination of EFB and cow manure with a ratio of 2:1 was the best composition of organic matter with the highest pH and the lowest dissolved heavy metals and sulfates in accordance with the established quality standards.*

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INTRODUCTION

Environmental problems due to the mining activities are the formation of acid mine drainage (AMD). The formation of AMD in mining activities is the impact of the exposure of mineral rocks containing pyrite (FeS_2). Acid mine drainage is formed by hydro-geochemical weathering of sulfide rocks (marcasite, arsenopyrite, and pyrite) in contact with oxygen and water. This reaction is catalyzed by sulfur and iron-oxidizing microorganisms (Aksel and Kaldas 2006). High heavy metal content, very acidic pH, and high sulfate levels seriously threaten groundwater and surface water pollution (Rambabu et al. 2020).

AMD formation has been prevented by minimizing the contact between oxygen, water, and sulfide minerals, such as encapsulation, storage of tailings waste below the water's surface, and application of anionic surfactants (Johnson and Hallberg 2005; Kuyucak 2002). Meanwhile, the treatment of AMD currently being carried out in various countries, including Indonesia, is by active and passive treatment (Rambabu et al. 2020), depending on the input provided. Active treatment is carried out with the addition of lime and alum to increase pH, remove metals and sulfates, and reduce TSS from wastewater, but in the long term will produce gypsum deposits (Masindi 2017) which require periodic dredging. Active treatment costs are also very high when contaminants are high in volume and require continuous addition and monitoring of materials (Ruehl and Hiibel 2020). Another alternative is a passive treatment, which is an option because it requires little or no input during long-term operation, resulting in low costs and minimal maintenance. The passive treatment utilizes local materials that are always present in nature so that this system is cheaper and more efficient. Passive treatment relies on naturally occurring chemical and biological processes (Munawar 2007). The most commonly used type of passive treatment is wetlands (Pat-Espadas et al. 2018).

In the wetland system, biological, chemical, and physical processes occur naturally because of the interaction between plants, organic matter, and microorganisms in the environment. In anaerobic wetland systems, the matrix composition uses organic matter. The role of organic matter in the wetland system is as an SRB stimulator which can further increase pH and reduce sulfate and metal ions in water, as a metal chelating agent, and as a substrate to support plant growth (Chang et al. 2000; Sekarjannah et al. 2021). The application of organic matter could improve the quality of AMD by increasing the pH and decreasing the levels of dissolved Fe and Mn (Munawar 2007). Therefore, this study was conducted to select various types of organic matter available in nature that can improve the quality of AMD. In this study, there were 3 (three) stages of continuous research, namely, screening of a single type of organic matter, a combination of two different types of organic matter, and a combination of cow manure and empty fruit bunch (EFB) in various compositions.

METHODS

The application of organic matter is divided into several stages on an ongoing basis; there is the incubation of organic matter as an initial screening to obtain types of potential organic matter, then a combination of potential organic matter with organic matter that is abundantly available, and continued with testing the composition of the best organic matter.

Screening Types of Organic Matters

The research design used was a completely randomized design (CRD) with one factor the type of organic matter consisting of cow manure, goat manure, chicken manure, three types of compost, EFB, sawdust, wood chips, chopped water hyacinth, cocopeat, cajuputi leaf fresh waste, cajuputi leaf compost, waste of citronella distillation, baglog waste, and bagasse. The test was carried out by adding 100 grams of organic matter to 500 ml of AMD (Othman et al. 2015). The measured variable was pH which was observed every hour for 5 hours using a pH meter. The treatment was repeated three times so that there were 48 experimental units.

Combination of Types of Organic Matters

This study carried a combination of plant waste, such as wood chips, sawdust, bagasse, cocopeat, and EFB, with cow manure and chicken manure. This is done because plant wastes act as suppliers of lignin and cellulose for the long-term survival of sulfate-reducing bacteria (SRB), while cow manure and chicken manure act as carbon suppliers, which are used directly by SRB (Cocos et al. 2002). The test was carried out by adding 100 grams of organic matter to 500 ml of AMD (Othman et al. 2015). The measured variable was pH which was observed every hour for 5 hours using a pH meter. The research design used was a CRD in which each treatment was repeated three times so that there were 60 experimental units.

Combination of Cow Manure and EFB in Various Compositions

Based on previous research with the results that the ratio of EFB:cow manure (1:1) and EFB:cow manure (3:1) both were able to increase the pH consistently for 5 hours of observation. It is necessary to examine in detail the best composition of the combination of cow manure and EFB that could increase pH and reduce heavy metals in AMD effectively. The research design used was a CRD with three treatment levels with repeated three times so that there were 9 experimental units. The test was carried out by adding 100 grams of organic matters to 500 ml of AMD (Othman et al. 2015). The variables measured were pH, Fe, Mn, sulfate, and sulfide levels. The pH variable was conducted every hour for 5 hours of observation using a pH meter. While the content of Fe, Mn, sulfate, and sulfide was carried out before and after treatment.

Statistical Analysis

Data analysis used analysis of variance (ANOVA) at a level of 5%. If there was a significant difference, it was continued with the Duncan Multiple Range Test (DMRT) with a level of 5%. Data were analyzed using SPSS v16 Software.

RESULT AND DISCUSSION

Screening of Organic Matters

The changes in pH values due to tested organic matter are presented in Figure 1. The results show that adding certain organic matters can increase the pH from 3.05 to > 6 (Figure 1). This study showed organic matter of goat manure, EFB, compost, cow manure, baglog waste, chicken manure, chopped water hyacinth, waste of citronella distillation, and cajuputi leaf compost could increase AMD pH to > 6. The best result is that the addition of EFB can increase the pH to 8.07 with a retention time of 1 hour. Meanwhile, cocopeat, wood chips, sawdust, and cajuputi leaf fresh waste showed a slower pH increase and still < 6; even bagasse could only increase the pH to 4.

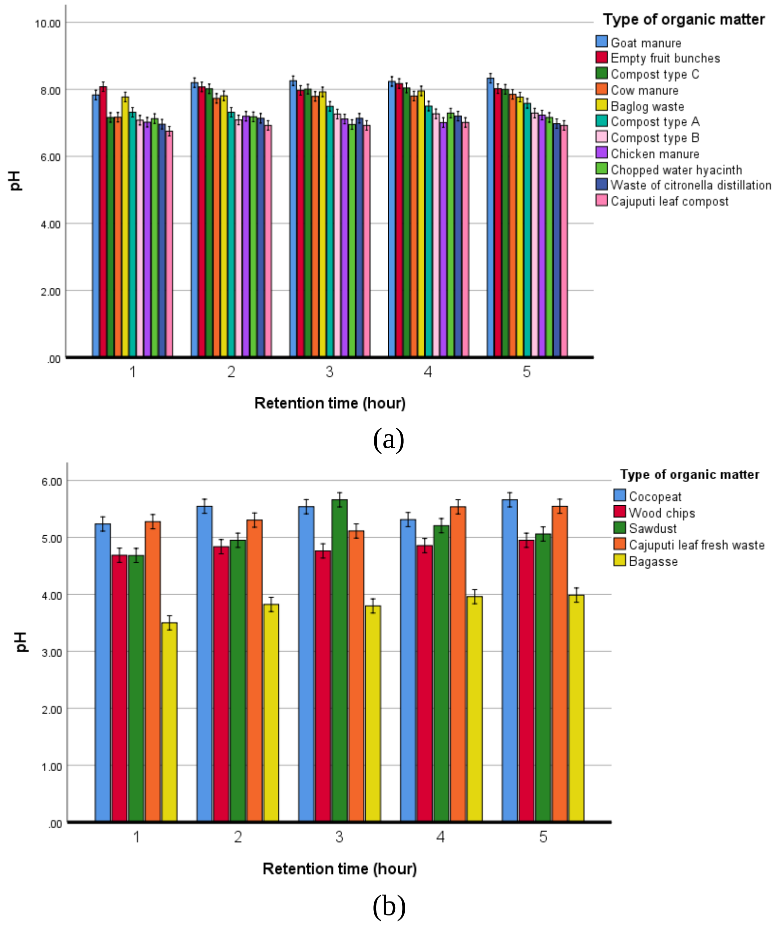

Figure 1 Effect of organic matter on AMD pH; (a) types of organic matter with a brief increase in pH, (b) types of organic matter with a rather slow increase in pH

The increase in pH occurs because organic matter can create reduction conditions due to the decomposition process of organic matter in the system (Headley and Tanner 2006). This condition causes the iron ion, which was initially ferric (Fe^{3+}), to be reduced to ferrous ion (Fe^{2+}) and releases one molecule of OH^- where the OH^- ion plays a role in increasing the pH. The increase in the pH of the water was very significant in the first hour and decreased after that. However, it was still stable, showing a neutral number because the decomposition process took place more quickly at the beginning (Effendi 2003).

$$Fe(OH)_3 + e^- \longleftrightarrow Fe(OH)_2 + OH^-$$

Types of organic matters such as cocopeat, wood chips, sawdust, fresh waste of cajuputi leaves, and bagasse can increase the pH more slowly than other types. This is because these types of organic matter have high lignin content and are still very fresh, so the decomposing microorganisms need a long time to decompose. It takes a long time to create reduction conditions in the system so that the pH increase is slower. From this research, it was found that the type of organic material that was able to increase pH, abundant in nature, and cheap, is EFB; then, types of organic matters that can increase pH, their availability is more limited and expensive, such as cow manure and chicken manure; and types of organic matters that are slow in increasing pH but are inexpensive and abundantly available in nature, such as sawdust, wood chips, cocopeat, and bagasse.

Combination of Organic Matters

The combination of types of organic matter resulted in a different increase in pH for each treatment. This study carried a combination of plant waste, such as wood chips, sawdust, bagasse, cocopeat, and EFB, with chicken manure (Figure 2) and cow manure (Figure 3). In selecting organic matters for AMD remediation, it is necessary to pay attention to their availability around the field, such as the principles of SDGs (Sustainable Development Goals) are efficiency, accessibility, and community participation, so it is hoped that the materials used do not come from outside the region. Moreover, it can simultaneously apply the circular economy principle, utilizing waste and extracting the maximum value from using these materials. Therefore, this research combines types of organic matters that can rapidly increase pH and organic matters that are abundantly available.

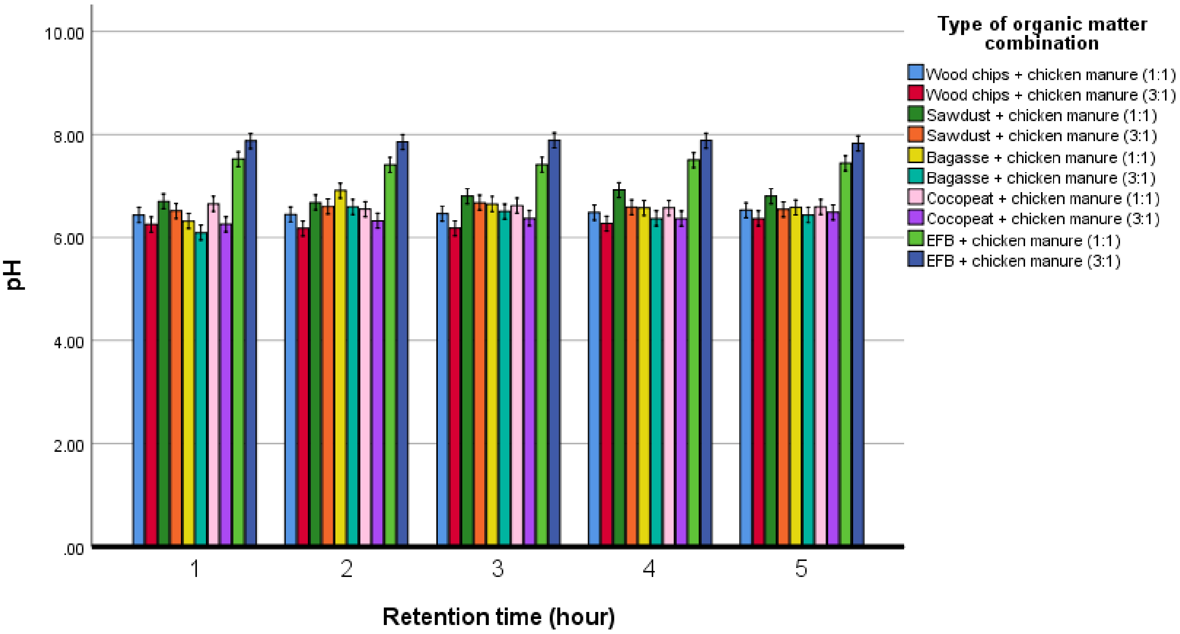

Figure 2 Effect of combination of organic matter (plant waste and chicken manure) on AMD pH

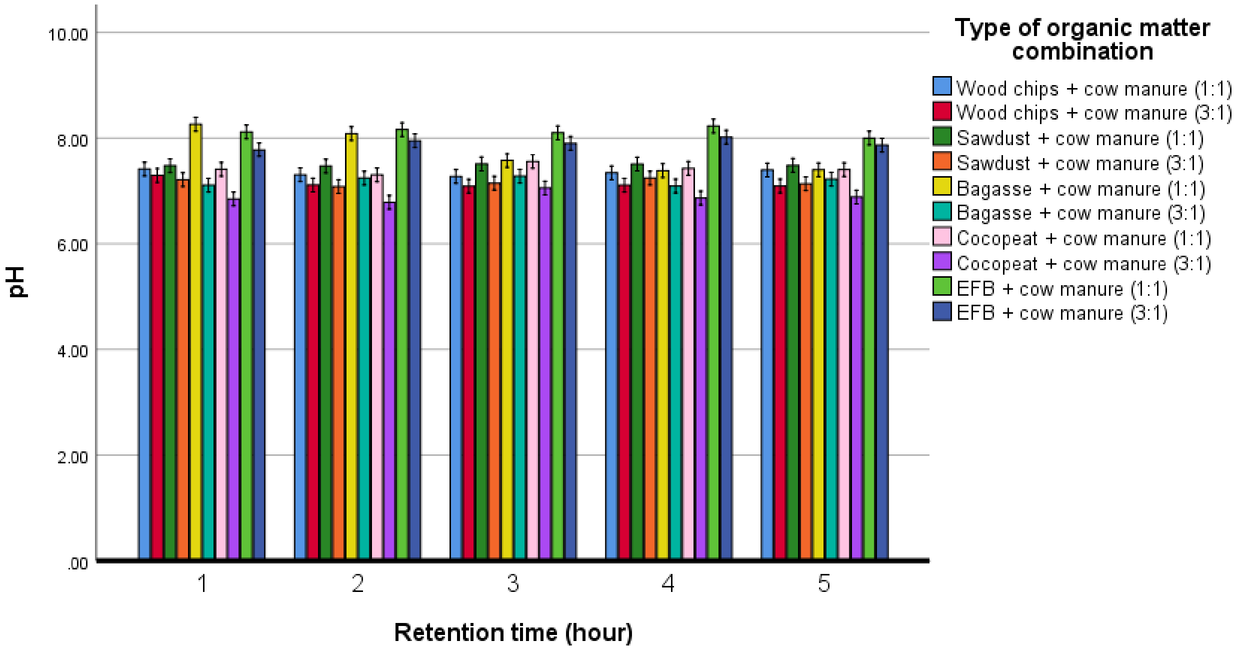

Figure 3 Effect of combination of organic matter (plant waste and chicken manure) on AMD pH

The results showed that all the treatments tested could increase the pH from 3.26 to > 6 for 1 hour. However, the overall increase in pH by combining plant waste and chicken manure (Figure 2) was lower than the increase in pH by combining plant waste and cow manure (Figure 3). The treatment with the highest increase in pH was a combination of EFB and chicken manure in a ratio of 1:1 and 3:1 (Figure 2). The treatment with the highest increase in pH in the first hour was a mixture of bagasse and cow manure (1:1) but decreased with the length of time observed (Figure 3). This is because the organic matter that has good performance in increasing the pH of AMD is only cow manure, while bagasse requires a longer time, so a significant increase in pH only occurs in the first 1 hour. It is different with the combination of EFB and cow manure in a ratio of 1:1 and 3:1, which can increase the pH from 3.26 to 8.12 and 7.78, respectively, with a retention time of 1 hour and consistent for 5 hours with a pH between 7.78–8.23. The statistical analysis results showed that the two treatments were not significantly different ($P > 0.05$), so a more detailed study was carried out to determine the best composition of the combination of EFB and cow manure in increasing pH and reducing heavy metals and sulfate in AMD.

Combination of Cow Manure and EFB in Various Compositions

Analysis of the chemical characteristics of EFB and cow manure was carried out to determine the elements that play a role in the AMD quality improvement process, shown in Table 1. Empty fruit bunch and cow manure have a high pH and content of Ca, Mg, and K cations to increase the pH of AMD (Table 1). The organic matter with high pH and alkaline cations (Ca, Mg, K, and Na) has the potential to be a source of alkalinity for AMD (Munawar and Riwandi 2010), which is consistently able to increase AMD pH > 7 (Gibert et al. 2004). In addition, EFB and cow manure also have high cation exchange capacity (CEC) values, 29.36 and 63.22 cmolc/kg, respectively, where a high CEC indicates that the material can adsorb or exchange cations, in this case, able to remove dissolved metal cations in AMD (Munawar and Riwandi 2010). Empty fruit bunch can be used as a natural adsorbent for heavy metal removal because it has a large active surface area of 53.3 m^2/g. The active surface area plays an essential role in the heavy metal adsorption process (Khosravihaftkhany et al. 2013).

Table 1 Characteristics of empty fruit bunch and cow manure

Parameter	Empty fruit bunch	Cow manure
pH	9.1	8.8
C-organic (%)	54.43	41.72
Total N (%)	1.19	1.54
C/N ratio	46	27
Total P (%)	0.06	1.79
Total K (%)	3.58	7.17
Total Ca (%)	0.6	3.23
Total Mg (%)	0.47	1.02
CEC ($cmol_c$/kg)	29.36	63.22

The provision of EFB and cow manure is a source of energy needed by SRB, where SRB will play a role in reducing sulfate content and decreasing heavy metal content through the formation of metal sulfides (Hards and Higgins 2004). Empty fruit bunch and cow manure have C/N ratios of 27 and 46, while the optimal C/N ratio favored by SRB is 45–120 (Gibert et al. 2004), but a C/N ratio of 20 is also capable of initiating the growth of SRB (Munawar and Riwandi 2010). Organic matters with low C/N supply carbon can be used directly by SRB. In contrast, organic matters with high C/N ratios, such as EFB will ensure the need for energy sources for SRB in the long term. The mixture of biodegradable materials with materials that can be degraded in the long term will increase the efficiency of SRB survival in the long term (Willquist et al. 2015). Therefore, EFB and cow manure used in this study is the right combination.

Prior to the start of the study, an AMD quality analysis was carried out, aiming to determine the difference in the quality of AMD before and after treatment. The results of the analysis are shown in Table 2. From the analysis of the quality of AMD, it was obtained that AMD had a very low pH of 2.17 and high solubility of heavy metals Fe, Mn, and sulfate at 31 mg/L, 8 mg/L, and 394 mg/l, respectively. The sulfide content value is still very low, < 0.01, indicating no hydrogen sulfide (H_2S) has formed yet. The pH value and dissolved heavy metals Fe, Mn still do not meet the quality standards of the Minister of Environment Decree No. 113 of 2003 concerning Wastewater Quality Standards for Coal Mining Business and or Activities. Therefore, it is essential to manage AMD, one of which is the addition of organic matter.

Table 2 The quality of AMD before treatment.

Parameter	Acid mine drainage*	Quality standard**
pH	2.17	6–9
Fe (mg/L)	31	7
Mn (mg/L)	8	4
Sulfate (mg/L)	394	-
Sulfide (mg/L)	< 0.01	-

*: *Services Laboratory* SEAMEO BIOTROP; **: Minister of Environment Decree No. 113 of 2003 concerning Wastewater Quality Standards for Coal Mining Business and or Activities

There are three treatments in this research, namely A1 (combination of EFB:cow manure 1:1), A2 (EFB:cow manure 2:1) and A3 (EFB:cow manure 3:1). The results showed that the pH of AMD, which was 2.17 increased to 7.27 in A1 treatment, 7.55 in A2 treatment, and 7.58 in A3 treatment for 1 hour and increased with the length of time observed (Figure 4). The A1 treatment was significantly lower ($P < 0.05$) than the other treatments from the statistical analysis. In contrast, the treatment of A2 and A3 showed that the results of increasing pH were not significantly different ($P > 0.05$).

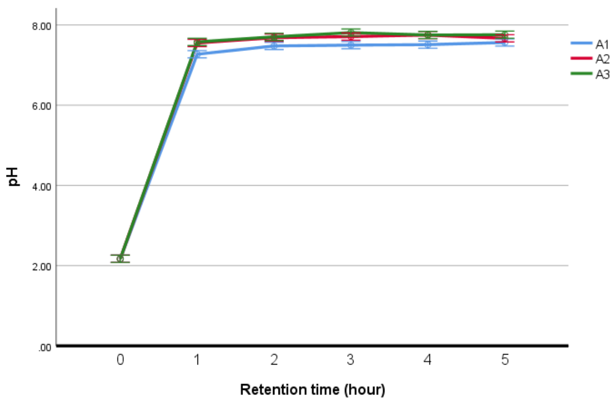

Figure 4 Effect of composition of EFB and cow manure on AMD pH

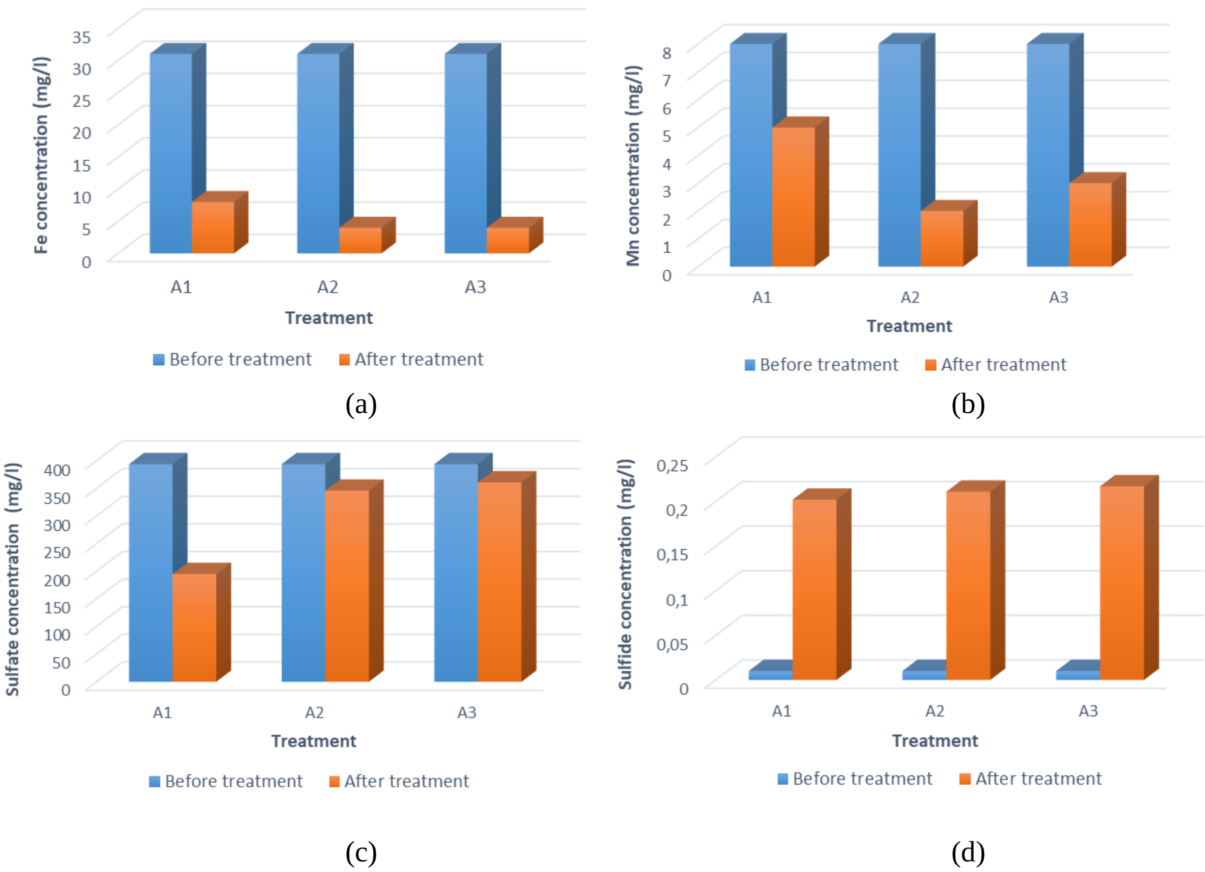

Figure 5 The effect of the composition of EFB and cow manure on (a) Fe concentration; (b) Mn concentration; (c) Sulfate; (d) Sulfide in AMD

Analysis of heavy metals Fe and Mn, sulfate, and sulfide content was also carried out to see the effect of organic matter application on AMD quality (Figure 5). From Figure 5, it can be seen that at a retention time of 5 hours, the combination of EFB and cow manure reduced heavy metal Fe in treatment A1 up to 8 mg/L, treatment A2 and A3 up to 4 mg/L. For the dissolved Mn, it was found that in treatment A1 decreased up to 5 mg/L, in treatment A2 up to 2 mg/L, and in treatment A3 up to 3 mg/L. For treatment, A1, the dissolved Fe and Mn were still above the specified quality standard threshold, while treatments A2 and A3 were already below the environmental quality standard threshold. The addition of organic matter could also reduce the dissolved sulfate in AMD. In treatment A1 decreased to 195 mg/L, treatment A2 to 346 mg/L, and A3 to 361 mg/L. Simultaneously with the decrease in the dissolved heavy metals and sulfates, the sulfide content in the water increased due to the sulfate reduction process occurring in the system by 0.202; 0.211; and 0.217 mg/L in treatments A1, A2, and A3, respectively. Determination of the best organic matter composition in acid mine water management can be seen in Table 3.

Table 3 The effectiveness of organic matter on AMD quality on various parameters

Parameter	Treatment		
	A1 (EFB:cow manure 1:1)	**A2 (EFB:cow manure 2:1)**	**A3 (EFB:cow manure 3:1)**
pH	7.27	7.55	7.58
Fe removal (%)	74.2	87.1	87.1
Mn removal (%)	37.5	75	62.5
Sulfate removal (%)	50.5	12.2	8.6
Sulfide rate (mg/l)	0.202	0.211	0.217

The best composition of organic matter was EFB and cow manure in a ratio of 2:1, because able to increase the pH from 2.17 to 7.55 in 1 hour and increased with the length of time observed (Figure 4). This combination was able to reduce the sulfate and heavy metal of Fe and Mn followed by 12%, 87%, and 75%, respectively, within 5 hours, and increased sulfide by 20% (Figure 5) as a result of the sulfate reduction process that occurred in the system. This research supports that the use of 300 tons of empty fruit bunches and 120 tons of compost as pond bottom substrate in an artificial swampy forest with an area of 3300 m^2 and a capacity of 1,500 m^3 can neutralize pH at a value of 6–7 during 4 hours incubation (Yusmur et al. 2019).

The role of organic matter in reducing sulfate and heavy metals is a stimulator of SRB. The reduction condition caused by the addition of organic matter supports the growth of SRB, which plays a role in reducing sulfate. Sulfate-reducing bacteria can use sulfate, sulfite, or thiosulfate ions as electron acceptors to obtain energy in their metabolic processes, while the organic matter is as electron donors (Hards and Higgins 2004). SRB metabolism requires low molecular weight organic compounds such as acetate as a carbon source. An essential mechanism for SRB activity is the sulfate reduction process under anaerobic conditions and optimal environmental conditions for sulfide formation. The following is the reaction for the formation of sulfide and the sulfate reduction process, which then reacts with metal cations to form metal sulfide (Drury 1999):

$$CH_3COO^- + SO_4^{2-} + H^+ \longrightarrow H_2S + 2HCO_3^-$$
$$Me^{2+} + H_2S \longrightarrow MeS + 2H^+$$

Where Me^{2+} represents cationic metals, such as tin (Pb), cobalt (Co), cadmium (Cd), copper (Cu), nickel (Ni), iron (Fe), manganese (Mn), and zinc (Zn). The sulfate reduction reaction consumes SO_4^{2-}, resulting in H_2S and bicarbonate (HCO_3^-), which are released, increasing the alkalinity of AMD. The resulting H_2S will react to form metal sulfides which precipitate and reduce heavy metals and sulfates from water (Drury 1999; Zagury et al. 2006). The decrease in dissolved heavy metals can also occur directly, where the metal will precipitate at a neutral pH (Gibert et al. 2004). Some metal elements, such as Fe, will soon be precipitated as (oxy) hydroxide (Gibert et al. 2004). This deposition is a characteristic of each metal and will occur at the beginning of incubation, even before the role of SRB (Amos and Younger 2003). Judging from the research results that the sulfate in the water is still high and the sulfide formed is still very low (Figure 5), the decrease in heavy metals is thought to have occurred due to a significant change in pH so that the metal itself will settle immediately. A metal-sulfide precipitation reaction controls the sulfide, with the dissolved sulfide increasing only after the metal in the water decreases (Cocos et al. 2002). It can be seen in Figure 5 that the sulfide in treatments A2 and A3 was higher than in treatment A1 because the dissolved metal in treatments A2 and A3 was lower than in treatment A1.

Referring to the notion of wetlands as productive habitats with anoxic waterlogged soils (Terry and Banuelos 1999), which can improve wastewater quality, vegetation, organic substrates, and microorganisms are the main components of artificial wetlands. The selection of organic matter is not only seen from its ability to provide carbon for SRB but also needs to consider its maturity to become a growth medium for plants to be planted. In terms of chemical properties, organic matters that are good for use as fertilizer for plants are those that contain C-organic 15, C/N ratio ≤ 25, and macronutrients (N+P+K) 2 (Ministry of Agriculture 2019). From the characteristics of EFB and cow manure used, the C-organic and macro-nutrient requirements have met the requirements, but the C/N ratio of EFB is still too high. This can be overcome by mixing EFB and cow manure. The combination of EFB and cow manure with a ratio of 2:1 is the best treatment to improve AMD quality. In addition, the ratio of 2:1 was chosen because it can reduce the C/N of the substrate due to its maturity as a growing medium and the high C-organic and macronutrient content for phytoremediation AMD. It is also relatively cheaper when viewed from a cost perspective, so this composition of organic matter is suitable for large-scale AMD management, especially in Indonesia. Indonesia has an area of 2.82 million hectares of oil palm plantations with a yield of 48.4 million tons (Central Bureau of Statistics 2019), where each 1 ton of palm oil will produce waste in the form of 23% empty fruit bunches or 230 kg. This is an abundant source of organic matter to deal with AMD problems in Indonesia. The utilization of organic matter seems to be promising in AMD treatment technology for improving environmental quality.

CONCLUSION

Several types of organic matter, including goat manure, empty fruit bunch, compost, cow manure, baglog waste, chicken manure, chopped water hyacinth, citronella distillation waste, and cajuputi leaf compost waste, were able to neutralize AMD. The composition between EFB and cow manure in ratio 2:1 is the best choice that can increase AMD pH in a short time, reduce sulfate and heavy metals Fe and Mn by 12%, 87%, and 75%, respectively, and increase sulfide by 20% with a retention time of 5 hours. The combination of EFB and cow manure can improve the quality of AMD to meet the environmental quality standards set.

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