Save our soil from heavy metals (Pb and Cd) accumulation for rice growth

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Abstract. Rice intensification has been very successful in Indonesia and was even appreciated by FAO in 1985. However, despite the success, paddy soils are frequently polluted with Pb and Cd which are mostly derived from phosphate (P) fertilizers. Considering that the soils have been used for the long-term cultivation, Pb and Cd contents both in the soil and rice have exceeded the tolerance level in food as defined by WHO namely 2 and 0.24 mg kg^{-1}, respectively. The highest Pb and Cd were found in soils cultivated for 80 years in Musi Rawas, Indonesia with values of 20.56 mg kg^{-1} and 0.72 mg kg^{-1} for soil and 3.11 mg kg^{-1} and 0.29 mg kg^{-1} for rice, while the lowest was found at 20 years with values of 17.82 mg kg^{-1} and 0.26 mg kg^{-1}. Furthermore, the highest rice Pb and Cd were obtained at 80 years of cultivation with values of 3.11 and 0.29 mg kg^{-1}, respectively, while the lowest was found at 20 years with values of 2.35 mg kg^{-1} Pb and 0.15 mg kg^{-1} Cd. Therefore, these metals need to be managed immediately. One of the methods to detoxify heavy metals is by application of organic material. Through a chelating process, heavy metals are immobilized and not absorbed by rice. Based on the results, rice straw biochar and compost at a rate of 10 tons ha^{-1} separately reduced Pb and Cd in soil from 13.60 to 3.00 mg kg^{-1} and 13.60 to 5.98 mg kg^{-1} for Pb and 0.62 down to 0.29 mg kg^{-1} and 0.64 down to 0.42 mg kg^{-1} for Cd, respectively.
Keywords: Biochar, heavy metals, rice intensification, organic material, paddy soil, Pb, Cd and soil pollution

1. Introduction

Rice is a staple food of Southeast Asian countries, specifically Indonesia, which produced in different varieties on the wet and dryland. Several efforts such as the application of fertilizers are usually carried out to intensify rice growth and yield. Besides, marginal lands distributed widely in Indonesia need serious fertilization. However, excessive fertilization might lead to the pollution of the soil. In the 1980s, agricultural intensification programs were conducted in Indonesia to increase rice production. One example of the programs is the application of agrochemicals such as inorganic fertilizers and pesticides.

Continuous inorganic fertilization such as P fertilizer derived from natural phosphate or SP-36/TSP increases Pb and Cd contents in the soil [1]. The longer the age of land cultivation, the higher the Pb and Cd contents in soil and rice as shown in Tables 1 and 2. Furthermore, the soil age between 20 to 80 years of cultivation causes an elevation in the heavy metals concentration ranging from 17.82 to 20.56 mg kg^{-1} for Pb and 0.26 up to 0.72 mg kg^{-1} for Cd, respectively. Moreover, heavy metals in rice increased from 2.35 up to 3.11 mg kg^{-1} for Pb and 0.15 up to 0.29 mg kg^{-1} for Cd in the intensive farming system (IFS) of agriculture in Musi Rawas, South Sumatra, Indonesia [2]. Meanwhile, the limit of Pb and Cd in the food according to the World Health Organization (WHO) is 2 mg kg^{-1} and 0.24 mg kg^{-1}, respectively [2]. The rice intensive farming system located in Belitang, East Ogan Komering Ulu, South Sumatra, also contained 11.66 mg kg^{-1} of Cd [3].

The accumulation of these metals in the human body causes heavy diseases in the kidney, as well as cancer, osteoporosis etc. Pb and Cd allegedly originate from P fertilizer because SP-36 contains 11 mg kg^{-1} Cd and 67.100 mg kg^{-1} Pb. The high metal contents is due to the raw material used for P fertilizer namely natural phosphates [4]. The pollution of intensive paddy fields with Pb and Cd is a serious problem at the moment that needs urgent attention. According to [5], the accumulation of heavy metals in wetlands has potential impacts on changes in soil properties. More danger is expected when the heavy metal is concentrated in the roots, leaves, and grain. Another study showed that 22-24% of the total metal content in rice biomass was concentrated in grain [6]

Table 1. Content of Pb in paddy soils and rice [2]

Age of land for IFS (year)	Soil Pb (mg kg^{-1})	Rice Pb (mg kg^{-1})
0	1.20	-
20	17.82	2.35
40	19.48	2.40
60	20.46	2.86
80	20.56	3.11

The linear regression plot between soil and/or rice Pb with the age of soil cultivation showed that $Y_{soil\ Pb} = 11.88 + 2.02\ Ln\ (x)$, $R^2 : 0.956$, $Ln\ (Y)_{rice\ Pb} = 2.06 + 0.005\ x$, $R^2 : 0.928$. These two equations showed that the long-term use of soil cultivation for paddy growth increases Pb content in soil and rice. Hence, this heavy metal needs to be eliminated in the soil to make crop production safe for animals and humans.

Table 2. Content of Cd in paddy soils and rice [2]

Age of land for IFS (year)	Soil Cd (mg kg^{-1})	Rice Cd (mg kg^{-1})
0	0.05	-
20	0.26	0.15
40	0.32	0.16
60	0.39	0.21
80	0.72	0.29

The linear regression plot between soil and/or rice Cd with the age of soil cultivation showed that $Ln\ (Y)_{soil\ cd} = 0.17 + 0.02\ x$, $R^2 : 0.913$; $Ln\ (Y)_{rice\ Cd} = 0.11 + 0.011\ x$, $R^2 : 0.938$. These two equations showed the soil and rice Cd increases with the higher age of soil cultivation, therefore, this metal is very dangerous for human health and needs to be detoxified.

2. Source of Pb and Cd Contamination on Paddy soil

The intensive farming system is inseparable from the use of fertilizers and pesticides to increase crop production. The compounds used in the inorganic fertilizer contain heavy metals of Pb and Cd as shown in Table 3. These heavy metals might increase in the soil as the fertilizer usage becomes higher. Furthermore, the application of P fertilizers inadvertently adds Cd and Pb to the soil [7], while pesticides are also applied in the intensive farming system. In the UK for example, approximately 10% of pesticides contain Cu, Hg, Mn, Pb, and Zn. Lead arsenate is used in orchards for years to control some parasitic insects, while arsenic-containing compounds are also widely used to control cow lice and banana pests in New Zealand as well as Australia [8].

Table 3. Heavy metals contents of various inorganic fertilizers [9]

Heavy Metals	P fertilizer (mg kg^{-1})	N fertilizer (mg kg^{-1})
Arsenic	2-1.200	2.2-120
Boron	5-155	-
Cadmium	0.1-170	0.05-8.5
Cobalt	1-12	5.4-12
Cuprum	1-300	-
Mercury	0.01-1.2	0.3-2.9
Lead	40-4.000	-
Mangaan	0.1-60	1-7
Molybdenum	7-38	7-34
Nicel	7-225	227
Selenium	0.5	-
Uranium	0-300	-
Vanadium	2-1.600	-
Zinc	50-1450	1-42

The use of pesticides such as Antracol WP, Dithane M 45, and Buldox 25 EC which contain heavy metals for vegetable protection in Indonesia is very intensive, specifically in plants that have high economic value. Based on previous studies, 30-50% of total production costs are used for pesticides [10]. Meanwhile, the intensive use of pesticides tends to increase residues in soil and plants, even in the body of animals, fish, or other aquatic biotas. Farmers practicing the intensive farming system in the rice center of Musi Rawas, South Sumatra, Indonesia, use inorganic fertilizer including 166.37 to 192.85 kg ha^{-1} for Urea, 130.58 to 162.85 kg ha^{-1} for SP-36/TSP, 67.50 to 76.84 kg ha^{-1} for KCl and NPK fertilizer with a dosage of 123.70 up to 141.66 kg ha^{-1}. Usually, Indonesian farmers use P fertilizer with a dosage of 100 kg ha^{-1}, which is beyond the recommended limit.

3. Overcoming heavy metals of Pb and Cd in soil

According to [11], the stabilization of heavy metals in situ is performed by adding land amendments such as biochar and compost to reduce the bioavailability and minimize absorption by plants. Study conducted by [12] reported that the use of rice straw biochar on ultisol reduced the mobility of Pb and Cd, while [13] stated that the use of 40 tons ha^{-1} wheat branch biochar on Cd contaminated soil in the Jiangsu Yipeng area of China decreased Cd concentration in the plant and grain tissue. Furthermore, [14] used bivalve shell biochar and cow bone to reduce Pb toxicity up to 92.5%, and 75.8%, respectively, while [15] reported that biochar derived from garden waste at four levels of administration, namely 0%, 1%, 5%, and 10% in mine waste decreased the bioavailability of Cd, Pb, and Zn. [16] explained that biochar applications enhance immobilization of heavy metals in the soil, while [17] compared the use of biochar and compost to reduce Pb and Cd in the soil as shown in Tables 4 and 5.

Based on the results, rice straw biochar (RSB) and rice husk biochar (RHB) tends to effectively reduce Pb compared to rice straw compost (RSC) and rice husk compost (RHC). The highest reduction was found in the soil applied with RSB at the rate of 10 tons ha^{-1} with a value of 3 mg kg^{-1} (Table 4). Therefore, the application of Rice Husk Biochar (RHB) decreases the soil Cd and Pb with values ranging from 29.68% and 77.5%. Similarly, the application of RHB has high efficiency for reducing the solubility of heavy metals in rice with a value of over 80% ranging from 82.75-97.75%.

A similar result was found for soil Cd, namely the application of biochar reduced soil Cd more intensively compared to compost only. The application of rice straw and husk biochar at the dosage of 10 tons ha^{-1} reduced soil Cd with values of 0.29 and 0.30 mg kg^{-1} compared to the control with the value of 0.64 mg kg^{-1} (Table 5). Therefore, it was concluded that biochar has good potential to remediate soil heavy metals. As presented in Tabel 6, the interaction between biochar and compost decreases rice Pb and Cd below 0.07 mg kg^{-1} (Table 6), this is below the WHO recommended value for food tolerance limit of heavy metal contents with values of 2 mg kg^{-1} for Pb and 0.24 mg kg^{-1} Cd. Biochar as a soil enhancer derived from the burning of agricultural waste with limited oxygen has proven to have good potential as a soil enhancer since C-organic persist in black carbon and have long-term effects in chelating metallic elements [18]. Several other studies also showed that biochar application on soils increases upland rice yields [19] and rice crops on acid sulphate soils [20].

Table 4. Effect of biochar and compost on soil Pb after 60 days incubation [17]

Rates (ton ha^{-1})	Type of compost and biochar			
	RSC	RHC	RSB	RHB
	mg kg^{-1}			
0	13.60 i	13.60 i	13.60 i	13.60 i
2.5	5.04 ef	8.86 h	4.08 bcd	4.47 de
5	3.83 abc	7.36 g	3.64 abc	4.42 cd
7.5	3.31 ab	6.04 f	3.35 abc	3.29 abc
10	3.17 ab	5.98 f	3.00 a	3.06 ab

Note: Values in column followed by same small letter show insignificance difference at 5% Duncan test

Table 5. Effect of biochar and compost on soil Cd after 60 days incubation [17]

Rates (ton ha^{-1})	Type of compost and biochar			
	RSC	RHC	RSB	RHB
	mg kg^{-1}			
0	0.64	0.64	0.64	0.64
2.5	0.55	0.62	0.51	0.55
5.0	0.50	0.38	0.41	0.45
7.5	0.33	0.41	0.27	0.33
10	0.32	0.42	0.29	0.30

Note: Values at each column show insignificance difference at 5% Duncan test

Table 6. Effect of biochar and compost and its interaction on rice Pb and Cd contents [21]

Rate of Biochar	Rate of Compost			
	7.9 tons ha^{-1} RSC		6.88 tons ha^{-1} RHC	
	(mg kg^{-1})			
	Pb	Cd	Pb	Cd
7.24 tons ha^{-1} RSB	<0.04	<0.005	<0.04	<0.005
9.85 tons ha^{-1} RHB	0.07	0.005	0.06	0.005

The addition of organic ingredients and recycling actions provide great advantages. Typical compounds which contribute to the formation of complexes and ion exchange in organic matter are functional groups such as carboxyl (-COOH), hydroxyl (-OH), Carbonyl (= C = O), methoxyl (-OCH3), and amino (- NH2). One of the organic materials suitable for biochar materials is the rice husk and straw wastes which are widely available as local resources. Rice husk and straw biochar have different physical and chemical characteristics that allow the remediation and improvement of soil properties in intensive rice farming polluted by heavy metals. The use of biochar derived from bamboo leads to the absorption of Cu, Hg, Ni, and Cr both in soil and water, as well as Cd in contaminated soil [22], [23]. Meanwhile, the use of biochar derived from cow dung at a pyrolyzed temperature of 200°C is more effective in absorbing Pb than at 350°C due to the higher phosphate concentration lower

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temperature [24]. Furthermore, the use of rice straw biochar reduces the radical mobility of Pb, Cd on Ultisol [25], while others made from wood decreased the mobility of Zn and Cd up to 90% [26]. The use of cotton biochar to remove Cd contamination in the soil and absorption of Cd by cabbage reduced the bioavailability of Cd soil by adsorption or co-precipitation [27].

Rice straw is very abundant during the harvest season and is applicable as a compost material. The result of straw compost analysis with 3 weeks composting time includes C/N ratio 18.88, C 35.11%, N 1.86%, P_2O_5 0.21%, K_2O 5.35% and Water 55%. Based on these data, the straw compost has nutrient content equal to 41.3 kg urea, 5.8 kg SP36, and 89.17 kg KCl ton^{-1} of compost or total 136.27 kg NPK ton^{-1} of dry compost. Moreover, the results of [28] showed that the administration of straw compost for 3 planting seasons in a row produced no significant difference with the fertilization of SP-36 and KCl using a dose of 50 kg ha^{-1} each. This implies that there is no need for SP-36 and KCl fertilization on land that has been treated with inorganic straw compost for 3 seasons in a row. Meanwhile, the use of 5 tons ha^{-1} of straw compost in South Sumatera reduced the use of KCl fertilizer by 50% of the total requirement without the use of organic material [29]. Study conducted in West Sumatera Province on a newly opened paddy field also showed that the application of 2.5 tons rice straw ha^{-1} reduced KCl requirement from 100 kg ha^{-1} to 75 kg ha^{-1} and effectively increased grain yield. In addition, 10 tons ha^{-1} of rice straw is sufficient to negate the application of potassium fertilizer and the results obtained were not significantly different from 100 kg KCl ha^{-1}, but iron poisoning was effectively reduced [30]. According to [31], the increase of wetland rice productivity in Wawo Oru village, Palangga district, South Konawe regency of Southeast Sulawesi, is caused by the use of straw compost. The use of cow dung compost with straw up to 1500 kg ha^{-1} increased rice production and efficiency of SP-36 and KCl [32].

The organic matter applied to the soil eventually decomposes to produce organic acids such as humic and fulvic acid, which have an important role in compacted soil granulation. Decomposition products directly release the various nutrients needed by plants such as N, P, K, S, Ca and Mg which are previously bonded in the material. Indirectly, this tends to increase the soil pH value and the available P, considering that the organic acids of the decomposition might be in contact with Al, Fe, Mn and dissolve through the chelation process to form organic metal compounds. Study by [33] reported that the addition of organic materials suppressed Al-exchangeability from 16.60% to 27.70% followed by the increased availability of P, while [34] found that fulvic acid has a greater role than humic acid in the release of P elements in the soil. This is due to lower humic acid mobility as well as lower molecular weight and greater total acidity. These properties determine the ability to form complexes with existing cations, including metals.

Another study by [35] on the effect of manure on Cr uptake in peanuts showed that the use of manure and compost reduced the heavy metal content on the land. The combination of compost and manure at a dose of 30 tons ha^{-1} in a ratio of 1:3 and 15 tons ha^{-1} fertilizer in a ratio of 1:1 absorbed chromium metal from the soil up to 67 - 68%.

4. Mechanism of Organic matter in Remediating the Soil Heavy Metals

4.1. Effect of biochar on heavy metals concentration

According to [36], the mechanism of heavy metal adsorption by biochar is as follows: Pb^{2+} exchange with Ca^{2+} and Mg^{2+}, as well as other cations present in biochar, which leads to precipitation and innerphere complexation with humorous material complex as well as mineral oxides. Furthermore, complexation occurs with heavy metal surfaces and different functional groups such as free hydroxyl oxide minerals which precipitate on other surfaces followed by physical inperpheric adsorption and surface precipitation that contribute to Pb^{2+} stabilization. The general reaction between biochar and heavy metal is as follows:

$$[FG] + M^{z+} \longleftrightarrow [FM^{(z-1)}]^{+} + H^{+}$$

$$2\,[FG] + M^{z+} \longleftrightarrow [F_2M^{(z-2)}]^{+} + 2\,H^{+}$$

Where FG = functional grups (such as phenolic, carboxylate etc), M = metal with z valence, H = hydrogen.

Other components that play an important role include the presence of minerals such as phosphates and carbonates in biochar which bind and reduce the bioavailability of heavy metals in the soil [24]. Previous study by [37] stated that the primary mechanism of effective biochar to withstand heavy metals such as Pb is by precipitation of phosphate. Generally, during the manufacture of dissolved biochar, P, Ca, and Mg increases when heated to 200°C, but decreases at higher temperatures, this is likely due to an increase in Ca-Mg-P crystallization as evidenced by the formation $(CaMg)_3PO_2$ when the pyrolysis temperature increases to 500°C, thereby facilitating the precipitation of Pb.

The alkalinity of biochar also contributes to the precipitation of heavy metals in the soil. A study by [38] reported that the average pH value of biochar from various raw materials is 8.1. Furthermore, pH value increases with a rise in the pyrolysis temperature due to the ash content of the biochar. Therefore, most biochars are alkaline and have liming effects, which might contribute to the reduction of heavy metal mobility in the soil. Biochar derived from straw and rice husk decreased Pb by 77.94% and 77.5%, respectively [17].

4.2. Effect of compost on the heavy metal concentration

There are three mechanisms of heavy metal bonding with organic material (1) electrostatic interaction between metal and organic material, (2) formation of chelate compound between metals and functional groups which is different from humic fraction, as well as (3) formation of metal complex with humic fraction and water intermediate [39]. Organic matter functions as a regulator of heavy metal mobility in the formation of complex compounds and chelates with the metal. A complex is formed through the reaction between metal ions and ligands using electron pairs where ligands are called electron donors, while metal ions are called electron acceptors. Additionally, a chelate bond between organic and metallic materials might occur when the metal is bonded with a ligand at two or more bond points and form a ring structure, hence, the bond becomes more stable compared to the ligand bond at one point. Based on the results, composting from straw and rice husks tends to reduce Cd by 50% and 34% [17].

5. Conclusion

The intensive rice systems have been heavily polluted by heavy metals of Pb and Cd due to P fertilizers. Cultivated land for intensive agriculture contains high concentrations of Pb and Cd both in soil and rice. Meanwhile, efforts to overcome this problem include the application of amendments such as biochar and compost. Biochar is better at lowering Pb and Cd dissolved in soils, the application at 10 tons ha^{-1} decreased 78% Pb and 55% Cd, while compost of 10 tons ha^{-1} reduced Pb and Cd by 56-77% and 34-50%, respectively.

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