THE CONTAMINATION OF PB AND CD IN THE INTENSIVE PADDY FIELD AT MUSI RAWAS REGENCY, SOUTH SUMATERAINDONESIA

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Poll Res. 36 (4) : 1-6 (2017)

ISSN 0257–8050

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ABSTRACT

The farming activities in the intensive paddy field hadexcessively used theagrochemicals such as the phosphorous fertilizer and contributed to the increasing of heavy metal in the paddy field such as Lead (Pb) and Cadmium (Cd). The contamination poses serious problems due to affect the soil quality and agricultural products. This research aimed to investigate the accumulated of Pb and Cd in the soil and rice based on the utilization age of paddy field. The soil and rice samples were collected in the intensively paddy field used for 20, 40, 60 and 80 years in MusiRawas regency-South Sumatera-Indonesia. The concentration of Pb and Cd were measured by *Atomic Absorption Spectrophotometry* (AAS). The correlation between the heavy metal concentration and the utilization age of paddy field were analyzed using the linear regression test. The results showed that the increasing of the utilization age had suffered frommoderate to high heavy metal contamination. The highest concentration of Pb and Cd were at the utilization age of 80 years i.e. 20.56 ppm dan 0.72 ppm for the soil sample and 3.11 ppm and 0.29 ppm for the rice samples, andconcentration of Pb and Cd were at the utilization age of 20 years i.e. 2.35 ppm and 0.15 ppm for the rice samples. The results showed that the heavy metal contaminated the soil and rice had exceeded the maximum concentration regulated by WHO and need the further treatment to maintain the contamination.

KEYWORDS: Heavy metal contamination, Pb, Cd, Paddy field

INTRODUCTION

The paddy field in MusiRawasregency, in the South Sumatera, had been intensively utilized for agricultural activities for several decades. Because of the inappropriate management, the paddy field had degraded which characterized by physical, chemical, and biological damages.Furthermore, The paddy field could also be contaminated due to agrochemical, domestic, and industry wastes (Wuana *et al.*, 2011).

The increasing of inorganic fertilizer uses which was not seriously controlled by the use of adequate organic fertilizer could cause the increasing of heavy metal such as lead (Pb), and cadmium (Cd)which potentially contaminated the soil and the plants. The heavy metal contaminated soil might result in the important function losing of organic fertilizer to maintain the soil productivity which caused the physical, biological, and chemical damages. According to BalaiPenelitian Tanah (2010), there were about 65% from 8.1 million ha of paddy field in Indonesia poses the low organic molecule content (less than 2%).

The inorganic fertilizer especially phosphate (P) and nitrogen (N) fertilizer were categorized as the polluted source due to high heavy metal contained which endangered the agriculture land. For examples. The SP-36 fertilized contained 11 ppm of Cd and 67 ppm of Pb. The high level of Cd and Pb

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were caused by the raw industry material of phosphorous fertilizer, especially P from the natural phosphor which contained some metals and heavy metalssuch as Pb, Cd, Ca, Mg, Al, Fe, Si, Na, Mn, Cu, Zn, Mo, B, Hg, Cr, As, U, V, F, and Cl (Balai Penelitian Tanah, 2009).

The heavy metal contamination and the degradation of the intensive paddy field were the serious problems which needed the further treatment to maintain the problems. The accumulation of heavy metal in the agricultural land such as paddy field would influence the physical, biological, and chemical properties of soil (Satpathy*et al.,* 2014). In addition, the accumulation of heavy metal could also contaminate the roof, leaves, and grains which possessedrisks to the environmental safety and humanhealth if they distributed to the community as a food or the agricultural products (Singh *et al.*, 2011).Wang (2003) also reported that the plants have different abilities to absorb the contaminated heavy metal and the most distribution of heavy metal concentrated to the grain (22%-24%) (Wang *et al.*, 2003). This research aimed to investigate the contaminated heavy metal in the paddy field which had been used for several years (the utilization ages) and try to analyze the correlation between the amount of heavy metal and the utilization age of paddy field as the influences of the intensive fertilizer used.

MATERIALS AND METHODS

This study was conducted as the field studyand laboratory analysis. The field study was carried out to explore the contamination level of Pb and Cd as the heavy metal pollutant in the intensive paddy field. The exploration was based on the secondary data collection. The laboratory analysis was carried out in the Environmental Biotechnology Laboratory in Bogor to analyze the heavy metal concentration (ppm) and also the soil properties.

In order to determine the intensification of paddy field in MusiRawas regency, this research was begun by designing the questionnaire which given to the respondents in the four sub-district. They were Tugumulyo, Muara Beliti, Purwodadi and Megang Sakti sub-districts where were the center of rice production.The questionnaire was conducted to estimate how long the field had been utilized,kinds of the fertilizer, and pesticides, the drainage system, and the agricultural waste management.

The samples were selectedas the soil and rice which collected from four sub-districts. The additional chemical materials were 30% of hydrogen peroxide (H_2O_2), hydrochloride acid(HCl), sodium hydroxide (NaOH), ether, and the other chemical reagent to support the laboratory analysis. The supporting equipment were soil drill, tape meter, sieve, label, Atomic Adsorption Spectroscopy (AAS), Kjeldahl flask, analytical balance, analytical dropper, volumetric flask, volumetric glassware, Erlenmeyer flask, volumetric dropper, mortar and pestle, filter paper, oven, and the other equipment to support the laboratory analysis.

The soil sample was collected using soil drill at a depth of 0 to 20 cm at the harvest time. At each site, five samples were composited to form an individual composite sample. The samples were collected from the different utilization years of the intensive paddy field from 20, 40, 60, and80 years. The Pb and Cd concentration were conducted by measuring 1.00 g of the as-prepared sample and collected the sample to the digested tube. The samples were mixed using 1 ml perchlorate acid and 5 ml nitric acid and stored for one night. The sample was heated at 100°C for 1.5 h until the yellow gas had been fully run out. Furthermore, the samples were heated at 170°C for one hour and then heated again at 200°C for one hour until the white gas formed. The destruction of the sample was finished when the white precipitate formed, and the solution was approximately 1 ml. The solution was cooled to room temperature and diluted and mixed using distilled water to 10 ml. The solution was measured by AAS using the Pb and Cd concentration standard as the references standard.

The samples of rice were collected in the harvest period and consist of five sub-samples which composite to represent all the rice. The rice samples were collected from the intensive paddy field in the utilization ages of 20, 40, and 60 years. The Pb and Cd concentrations were determined using several procedures. The samples were collected at crucible and dried using furnace at 500°C until a constant weigh of the samples. The dried samples were meshed and stored in the dried and closed glassware. The samples were measured to 1 g and transferred to beaker glass and dissolved using distilled water to 50 mL. The samples were mixed using 5 mL and 2 mL of high concentrated $HNO_3$and HCl, respectively. The samples were heated at 100°C for 30 min and cooled to room temperatures. The sample solution was diluted using distilled water to 50 ml and filtered. The Pb and Cd concentration were measured by AAS. In addition, the analysis of Pb in the paddy was carried out using a descriptive method.

RESULTS AND DISCUSSION

Table 1 showed the Pband Cd concentration of as-prepared soil and rice based on the utilization age of the paddy field. Table 2 showed the simple linear regression between the utilization age of paddy field and the concentration of Pb and Cd in the samples. Table 3 showed the questionnaire results of the fertilizer consumption in five (5) sub-district in Musi Rawas Regency. Table 4 was the soil characteristics based on the utilization age of paddy field. Finally, Table 5 showed the questionnaire results of the utilization of leftover rice planting in five (5) sub-districts in Musi Rawas Regency.

The Pb and Cd Concentration in Soil

Table 1 showed the highest concentration of Pb and Cd in the soil found in 80 years of the utilization age i.e. 20.56 ppm and 0.72 ppm, respectively. In the other hand, the lowest concentration of Pb and Cd found in 20 years of the utilization age 17.82 ppm and 0.26 ppm, respectively.

The linear regression analysis showed 0.874 as the correlation coefficient which showed that the Pb concentration in soil had positively correlated to the utilization age of paddy field. The higher utilization age of the paddy land would generate the higher concentration of Pb in soil by the increase of percentage of 15.38%. Furthermore, the Cd concentration in soil also positively correlated to the utilization age of paddy field in which the correlation coefficient showed 0.928 and the increase of the percentage of 176.92 %.

The presences of heavy metal such as Pb and Cd were caused by the intensive use and high concentration of phosphorous fertilizer. The heavy metal which contained in the fertilizer could be accumulated as the increasing of the utilization age of paddy field. The questionnaire data showed that the farmer used the phosphate fertilizer in average i.e. 150. 26 kg $ha^{-1}$and it had exceeded the maximum dosage i.e. 100 kg ha^{-1}.

The Pb and Cd concentration in Rice

The highest concentration of Pb and Cd in rice found at 80 years of the utilization age i.e.3.11 ppm and 0.29 ppm, respectively (Table 1). In the other hand, the lowest concentration of Pb and Cd found at20 years of the utilization age i.e. 2.35 ppm and 0.15 ppm, respectively. The concentration of heavy metal had been exceeded the tolerance limit of Pb and Cd concentration regulated by World Health Organization (WHO) i.e. 2.00 ppm and 0.24 ppm, respectively. The correlation and regression analysis showed the positive correlation between the utilization age of paddy field and the Pb and Cd concentration. It was proved by the correlation coefficient of Pb and Cd analysis that showed 0.831 and 0.948, respectively. It could be concluded that the increasing of the utilization age of paddy field would positively increase the number of Pb and Cd concentration in the rice by the increase of the percentage of 32.34% and 93.33%, respectively.

The high amount of Pb and Cdin the paddy plant was carried out by the passive absorption. The passive uptake was also mentioned as the biosorption (Aprillia *et al.*, 2013). The biosorption was carried out by the absorption of the nutrient in the phosphorous fertilizer which added in the low fertility soil. The low fertility of soil would be supported by the fertilizer. Ali (2013) and Laghlimi *et al.* (2015) reported that the Pb and Cd were easily absorbed as their ion such as Pb^{2+} and Cd^{2+} by the

Table 1. Pb and Cd Concentration in soil and rice based on the paddy field year

Sample	Paddy field ages (year)	Pb		Cd	
		Concentration (ppm)	Deviation (%)	Concentration (ppm)	Deviation (%)
Soil	20	17.82	-	0.26	-
	40	19.48	9.32	0.32	23.08
	60	20.46	14.81	0.39	50.00
	80	20.56	15.38	0.72	176.92
Rice	20	2.35	-	0.15	-
	40	2.40	2.12	0.16	6.66
	60	2.86	21.70	0.21	40.00
	80	3.11	32.34	0.29	93.33

roots (Ali *et al.*, 2013 and Laghlimi *et al.*, 2015). The distribution of heavy metal was begun when the plant absorbed the nutrient as their metabolism. The contaminated heavy metal contained in the phosphorous fertilizer would be absorbed as the nutrients and translocated to the plant body and the rice.

The phosphorous fertilizer composition analysis resulted in the Pb and Cd concentration of TSP fertilizer as 120.60 ppm and 4.90 ppm, respectively. The SP-36 fertilizer contained Pb and Cd concentration as 5.3 ppm and 10.43 ppm, respectively. Rai (2013) explained that the accumulation of Pb and Cd in the soil were caused by the intensive use of phosphate fertilizer. In general, The phosphorous fertilizer contained P_2O_5as the micronutrient, but it also contained the dissolved heavy metal such as Pb and Cd which could threat in the intensive uses. (Aprilia dan Purwani, 2013). The presences of Pb and Cd in the phosphorous fertilizer were caused by the raw material of phosphate which made from the phosphate rocks. The phosphate rocks were mainly composed of phosphate and carbonate and a high amount of heavy metal such as Pb and Cd.

According to Rochayati (2012), the natural phosphate rocks given to the soil as the fertilizer would leave more than 90% of Pb and Cd in the soil (Rochayati *et al.*, 2012). The Pb and Cd could form as Pb^{2+} and Cd^{2+}. The presences of Pb and Cd in the soil could reduce the absorption of the nutrients due to high affinity to complex cationic exchange. Furthermore, the Pb and Cd could also dissolve in the soil, absorbed by the organic and inorganic colloid surface, highly bounded to the soil mineralization, and precipitated by the soil chemical compound, and contained in the living material.

The low contained C-organic i.e. 16.58 g kg^{-1} and 18.08 g kg^{-1} could increase the solubility and the absorption ability of Pb and Cd in the soil. It was caused by the important role of the organic compound as the regulatory of heavy metal mobility in the soil. The organic compound could form the complex molecule and form the chelate bounding with Pb and Cd. The complex molecules were formed as the donating of the pairing electron from the organic compound as the ligan to the heavy metal as the electron acceptor. The reaction was carried out using the Lewis acid-base reaction.

The ability of the organic compound to form the complex molecule with the heavy metal was caused by the high amount of functional group contained in the organic compounds.The functional group played the important role of the ability of organic molecule to form the complex molecule (Debela *et al.*, 2000; Wang *et al.*, 2003; Singh *et al.*, 2011; Liu *et al*, 2013; Satpathy *et al.*, 2014; Laghlimi *et al.*, 2015).

The analysis results showed that the pH soil of the research site had acid soil pH to the slightly acidic range between 5.56 and 5.08 (Table 4). The low soil acidity (pH) could increase the solubility of Pb and Cd in the soil and increased the absorption of the heavy metal. The soil acidity was the key factor to show the metal transformation. The soil acidity

Table 2. Calculation summary of simple linear regression model between the paddy field years and the concentration of Pb and Cd in soil and rice.

Independent Variables	Dependent Variables			
	Pb (soil)	Cd (soil)	Pb (rice)	Cd (rice)
The paddy field years	Y= -321.99 + 18.99X R square = 0.874	Y= -131.49 + 67.72X R square = 0.928	Y= 1.56 + 114.65X R square = 0.831	Y= -27.54 + 382.89X R square = 0.900

Table 3. The questionnaire summary of the fertilizer consumption in five (5) sub-district in MusiRawasRegency

No	Sub-district	Fertilizer (kg/ha)			
		Urea	SP36/ TSP	KCl	NPK
1	Tugumulyo	166.37	144.68	67.50	123.70
2	Megang Sakti	189.65	156.25	74.37	141.66
3	Purwodadi	181.94	162.85	76.84	131.57
4	Muara Beliti	191.27	156.90	64.54	133.22
5	Sumber Harta	192.85	130.58	69.23	131.42
	Total	922.20	751.26	352.48	661.57
	Average	184.44	150.26	70.49	132.31

played to control the chemical properties of Pb and Cd and the additional process in the soil (Taberima, 2004; Hidayati, 2013; Liu *et al.*, 2015). In general, the decreasing of soil acidity increased the presences of heavy metal such as Pb and Cd except for Mo and Se (Taberima, 2004).

The soil characteristic in the research site was shown in Table 4. The main fraction of soil was composed of sand as 69.24 and 73.41, dust as 18.26 and 23.20, and clay as 8.01 and 8.16. According to USDA triangle structure, the combination of the fraction ratio, the soil was included in the class of argillaceous sand. Jung (2008) and Liu (2015) reported that the increasing of fineness of the sand structure would increase the ability to absorb the heavy metals (Jung, 2008 and Liu *et al*., 2015). Therefore, the sand had the low ability to absorb the heavy metal compared to the clay.

According to the questionnaire data analysis, the farmer would utilize the leftover rice planting used as the compost (9.13%), cattle feed (20.43%), burned (55.22%), and leftover (15.22%) (Table 5). 90.87% of biomass as the residue of agricultural activities were not processedby converting the biomass such as straw, and husk as the organic source to increase the fertility of paddy field. The organic content in the research site was calculated as 16.58g.kg^{-1} to 18.08 g.kg^{-1}and could be improved by utilizing the biomass as the agricultural residue.

CONCLUSION

The Pb and Cd concentration in soil based on the utilization age of paddy field in MusiRawas regency were between 17.82 ppm to 20.56 ppm forPb, and 0.26 ppm to 0.72 ppm for Cd. The results showed that the increasing of the utilization age of paddy field positively increase the concentration of Pb and Cd in soil due to intensive use of the phosphorous fertilizer. In the other hand, the concentration of Pb and Cd in rice based on the utilization age of paddy field were in the range of 2.35 ppm to 3.11 ppm for Pb and 0.15 ppm to 0.29 ppm for Cd. The heavy metal concentration had excessed the maximum tolerance limit regulated by WHO i.e. 2.00 ppm for Pb and 0.24 ppm for Cd. The high concentration ofPb and Cd in soil and rice poses risks tothe environment safety and human well- being. The contaminated paddy

Table 4. Soil characteristics based on the paddy field years

Soil Characteristics	Age (years)				Unit
	20	40	60	80	
pH (H_2O)	5.56	5.52	5.50	5.46	-
pH (KCl)	5.08	5.12	5.11	5.09	-
C-organic	16.58	18.08	17.58	18.08	g kg^{-1}
P-available	14.10	13.50	11.40	13.80	mg kg^{-1}
K-dd	0.58	0.58	0.58	0.58	cmol(+)kg^{-1}
Na-dd	0.44	0.44	0.33	0.77	cmol(+)kg^{-1}
Ca-dd	1.10	1.50	1.48	2.08	cmol (+)kg^{-1}
Mg-dd	0.57	0.35	0.68	0.50	cmol (+)kg^{-1}
KTK	17.40	16.31	19.58	16.31	cmol (+)kg^{-1}
Texture					
- Sand	69.24	73.71	68.64	73.41	%
- Dust	22.75	18.26	23.20	18.47	%
- Clay	8.01	8.03	8.16	8.12	%

Table 5. The questionnaire results of the utilization of leftover rice planting in five (5) sub-districts in MusiRawasRegency

Sl. No	Sub-district	Utilization				Sub-total
		Compost	Cattle Feed	Burned	Leftover	
1	Tugumulyo	4	13	35	8	60
2	Megang Sakti	6	9	18	7	40
3	Purwodadi	2	10	23	5	40
4	Sumber Harta	6	8	18	8	40
5	Muara Beliti	3	7	33	7	50
	Total (%)	21 (9.13)	47 (20.43)	127 (55.22)	35 (15.22)	230 (100)

field needed the further management and treatment which could decrease the concentration of Pb and Cd using the integrated treatment. In addition, the utilization of biomass residue could be an alternative to decrease the use of phosphate fertilizer to support the plant nutrients.

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