Distribution pattern of heavy metals in soil fertilized with industrial sludge

Rozita Ahmad[A], Wan Rasidah Abdul Kadir[A] and Rosazlin Abdullah [B]

[A]Forest Plantation Programme, Biotechnology Forestry Division, Forest Research Institute of Malaysia (FRIM), 52109,Kepong, Malaysia, Email rozita@frim.gov.my, rashidah@frim.gov.my

[B]Department of Land Management, Faculty of Agriculture, Universiti Putra Malaysia (UPM), 43400, Serdang, Selangor, Malaysia, Email rosazlin@yahoo.com

Abstract

Bio-solids waste from pulp and paper mill industries have been linked to containing various metals as contaminants from the chemicals added during the pulping process. Metal pollution of soils is of great environmental concern especially when using paper mill sludge (pms) for land application. The aim of this study is to investigate the distribution of heavy metals in soil after being amended with composted and raw PMS as organic fertilizer for 14 months. A field plot on a *Tectona grandis* or Teak plantation was established at FRIM Research Station in Mata Ayer, Perlis by the application of different rates of composted and raw paper mill sludge (PMS). Inorganic fertilizer and control plot were set up to compare the effectiveness of the PMS as sludge fertilizer. Soil samples were collected at two depths of 0-15cm and 15-30cm from the surface, with three distances, 0.5m, 1.0m and 1.5m from the Teak trees. The distribution of Cadmium (Cd) in soil increased with depth for every distance. However there were no clear trends observed for Arsenic (As) and Lead (Pb) for different layers and distances. The concentration levels of As and Pb in soils were found to be higher than Cd for all the treatments, but the values were within the permissible range of the Maximum Permitted Concentration (MPC) for land application.

Key Words

Soil, heavy metals, sludge, composted paper mill sludge, teak.

Introduction

Pulp and paper manufacturing mill in Malaysia generates substantial amount of bio-solids from different stages of paper making. These waste residues are categorized under hazardous toxic materials by the Department of Environment, Malaysia (DOE). The current practices of sludge disposal such as land spreading, landfilling and incineration are becoming unfavourable due to environment and economic considerations (Krigstin 2005). Safe management and environmental friendly approaches must be developed to utilize these waste materials. PMS can provide beneficial attributes to soil ecosystem and crop yield due to its physical properties and good plant nutrient content. Through composting, paper mill waste can also be biodegraded into organic soils. The purpose of this study is to determine leaching of heavy metals in soil after 14 months application with composted and raw PMS.

Materials and Methods

Raw and Composted Paper Mill Sludge

Raw paper mill sludge (RPMS) was collected from the biological treatment pond from paper mill in Selangor, Malaysia. The material obtained was wet with approximately 75% moisture content, sticky, compact, caking in nature and have strong unpleasant smell. Dried shredded oil palm fibers were added as enhancement material to provide and improve aeration of the sludge. Pile composting method was applied with ratio of 1:1 of sludge and oil palm fiber mixture and evenly turned for the production of composted paper mill sludge (CPMS). This final product has soil like structure with organic soil colour, friable with no unpleasant smell. Table 1 provides the properties of raw and composted PMS.

Field Plot

The study plot was established at FRIM Research Station in Mata Ayer, Perlis, Malaysia. The soil in site has pH of 6.62 with 0.12% N and 1.00 % C content (Rosazlin *et.al.* 2008). A total of 480 *Tectona grandis* seedlings were planted at 3m by 4m distance. Eight treatments were carried out and described in Table 2.

Soil Sampling and Chemical Analysis

Soil samples were taken using soil auger at two depths 0-15cm and 15-30cm from the surface, with three distances, 0.5m, 1.0m and 1.5m from Teak plants. This is to represent the application area, soil underneath and surrounding of the treated site. Heavy metal elements in the soil were extracted using microwave digestion procedure followed the EPA 3051A method and concentration analysed on Varian 725 ICP-OES spectrometer. Duncan New Multiple Range Test (DMRT) was carried out to analyse the significant difference of each metals between the treatments. A one way ANOVA statistical analysis was also applied to test the significant difference in concentration of the elements between the depth and distance.

Table 1. Properties of composted and raw paper mill sludge.

Parameter	Composted PMS	Raw PMS	Maximum Permitted permission (MPC)
pH	4.74	7.84	
Organic C %	10.02	13.9	
Total N %	1.48	4.84	
C/N ratio	6.77	2.90	
P %	0.56	0.78	
K %	0.80	0.42	
Ca %	0.93	0.53	
Mg %	0.25	0.15	
Cu ppm	88	70	1,000 – 1,750
Zn ppm	188	153	2,500 – 4,000
Mn ppm	91	111	
Pb ppm	114	155	750 – 1,200
Cd ppm	0.21	0.29	20 – 40
Fe ppm	0.30	0.11	
Al ppm	0.88	0.90	
As ppm	8.07	15.54	

(Source: Wan Rashidah *et.al.*,2004)

Table 2. Application treatment carried out.

Treatment	Application per plant	Treatment	Application per plant	Treatment	Application per plant
T1	Control	T3	20 kg N/ha CPMS	T6	40 kg N/ha RPMS
T2	50 kg N/ha Inorganic Fertilizer	T4	40 kg N/ha CPMS	T7	80 kg N/ha RPMS
		T5	60 kg N/ha CPMS	T8	120 kg N/ha RPMS

Result and Discussion

Distribution patterns of heavy metals between treatments

Concentration of As, Pb and Cd in soil at different depths and distances after 14 months of applying composted and raw PMS are presented in Tables 3, 4 and 5 respectively. Applications with inorganic fertilizer contribute the highest concentration of As compared to other treatments except at 1.0 m distance. The highest As detected at 1 m distance is obtained from application of 60 kg N/ha and 40 kg N/ha of composted PMS at 0-15 cm and 15-30 cm depths respectively. Further away from the sampling point which was at 1.5 m distance, high content of As was observed from treatment with 20kg N/ha of composted PMS at 15-30 cm soil layer compared to control and other sludge application. However, at the same distance, there is no significant difference between treatments for As at 0-15 cm soil layer. The results show that high amount of As leached out into the soil are obtained from composted PMS treatment. This could be attributed to the inorganic fertilizer added into the composted PMS treatment.

Lead values are found to be higher in both soil layers at 0.5m distance for all the sludge treatments compared to control except for application with 80 kg N/ha raw PMS. Further away from the sampling site in 15-30 cm soil layer at 1.0 m distance, higher Pb content is observed for all application with composted and raw PMS compared to control and inorganic fertilizer treatment. No significant difference was found between treatments at soil depth of 0-15 cm for 1.0 m and 1.5 m distance. At 1.5 m distance, highest Pb content was detected from application with 40 kg N/ha of composted PMS at 15-30 cm soil horizon. The results indicated that there is a release of Pb from all the sludge application in soil depth of 15-30 cm soil at 1.0 m distance after 14 months treatment but the concentration are within the accepted MPC range. Addition of organic admendments containing a proportion of organic matter can reduced the mobility of heavy metals through formation of insoluble organic matter and metal complexes. On the other hand, these metal complexes can become soluble releasing the metals into the soil at high pH values (Killbride 2004). In this investigation, there was no significant difference of Cd in soil between any of the treatments when the results were compared to control which has no sludge and fertilizer treatment. This suggested that application of PMS did not lead to an increase in Cd content in soil.

Table 3. As content in soil taken at different depths and distances after 14 months application with composted and raw PMS.

Treatment	0.5 m distance		1 m distance		1.5 m distance	
	0-15 cm	15-30 cm	0-15 cm	15-30 cm	0-15 cm	15-30 cm
T1- control	7.216 b	10.217 bc	8.074 ab	10.624 abc	8.183 a	9.811 b
T2- inorganic fertilizer	13.078 a	14.079 a	11.312 a	12.549 ab	11.675 a	15.234 a
T3 – composted PMS	6.756 b	9.493 bc	7.269 b	10.989 abc	9.151 a	11.454 ab
T4 – composted PMS	8.507 ab	7.175 c	5.759 b	13.586 a	7.243 a	7.179 b
T5- composted PMS	4.265 b	10.725 abc	11.397 a	9.832 abc	9.660 a	9.947 b
T6 – raw PMS	8.677 ab	10.910 ab	8.341 ab	9.851 abc	7.636 a	10.045 b
T7 – raw PMS	6.540 b	8.874 bc	6.313 b	6.591 c	9.616 a	8.375 b
T8 – raw PMS	6.433 b	8.115 bc	7.594 ab	7.990 bc	7.932 a	7.723 b

Table 4. Pb content in soil taken at different depths and distances after 14 months application with composted and raw PMS.

Treatment	0.5 m distance		1 m distance		1.5 m distance	
	0-15 cm	15-30 cm	0-15 cm	15-30 cm	0-15 cm	15-30 cm
T1- control	6.339 ab	7.354 b	8.955 a	8.094 b	9.805 a	12.209 ab
T2- inorganic fertilizer	6.218 ab	13.631 a	6.449 a	7.733 b	2.565 a	6.239 c
T3 – composted PMS	9.431 a	9.245 ab	8.880 a	13.385 a	9.741 a	10.520 ab
T4 – composted PMS	8.789 a	9.709 ab	9.102 a	9.222 ab	8.661 a	12.814 a
T5- composted PMS	10.349 a	9.796 ab	10.426 a	11.600 ab	8.542 a	11.773 ab
T6 – raw PMS	9.381 a	10.324 ab	10.174 a	9.593 ab	7.203 a	7.958 bc
T7 – raw PMS	3.894 b	7.302 b	6.845 a	9.671 ab	7.939 a	8.159 bc
T8 – raw PMS	8.192 a	10.723 ab	7.727 a	8.196 ab	10.507 a	8.301 bc

Table 5. Cd content in soil taken at different depths and distances after 14 months application with composted and raw PMS.

Treatment	0.5 m distance		1 m distance		1.5 m distance	
	0-15 cm	15-30 cm	0-15 cm	15-30 cm	0-15 cm	15-30 cm
T1- control	3.628 a	5.499 a	4.083 a	5.815 a	4.495 a	5.300 a
T2- inorganic fertilizer	4.804 a	5.707 a	3.673 a	4.604 a	3.471 a	4.741 a
T3 – composted PMS	3.991 a	4.398 a	3.716 a	4.432 a	3.364 a	4.398 a
T4 – composted PMS	3.929 a	5.533 a	4.127 a	5.929 a	4.207 a	5.716 a
T5- composted PMS	4.542 a	5.651 a	5.235 a	5.923 a	4.499 a	5.563 a
T6 – raw PMS	4.297 a	6.093 a	4.328 a	5.310 a	3.988 a	5.449 a
T7 – raw PMS	4.748 a	5.804 a	4.706 a	5.705 a	4.737 a	4.934 a
T8 – raw PMS	4.036 a	5.277 a	3.983 a	4.756 a	4.096 a	4.161 a

Letter with the same alphabet list in the column for the different treatments are not significant different at $p<0.05$, according to the Duncan New Multiple Range Test (DMRT)

Distribution trends of heavy metals between soil depth and distance

Analysis of Variance tabulated in Table 6 indicated significant difference for Cd in soil between the two soil layers and distances. Concentration of Cd is observed to be higher in 15-30 cm soil layer compare to the upper soil horizon for all treatments applied. The distribution of Cd in soil increased with increasing depth for every distance. But Cd content did not show any clear trends with increasing distances.

No significant difference is observed for As and Pb concentration between the two soil layers and distances. These two elements did not exhibit any obvious pattern on the distribution in different depths and its surroundings. However the concentration levels of As and Pb in soils were found to be higher than Cd for all the treatments but the values did not exceed the Maximum Permitted Concentration (MPC) for land application.

Table 6. Statistical variation (Anova) for As, Cd and Pb between the two soil layers and distances.

Metal	*F*	*P-value*	*F crit*	*P-value* < 0.05 indicate significant difference
As	1.96	0.105	2.44	Not significant
Pb	1.43	0.235	2.44	Not significant
Cd	10.89	9.21E-07	2.44	Highly significant

Conclusion

The data obtained in this study demonstrate that application of PMS did not lead to an increase in heavy metals concentration in soil. There were no clear pattern to relate the leaching of metals to the lower soil layer and its surrounding.

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