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Effect of different dose of fly ash with and without FYM on heavy metals status in soil and plant parts of rice

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Abstract
The present study was carried out to evaluate the accumulation of heavy metals in soil and in paddy crop that have been amended by different dose of fly ash (20, 40, 60 t ha^{-1}) with and without FYM. In this study, the amount of heavy metals (Ni, Co, Cr, Pb and Cd) present surface and subsurface soils and various parts of the rice plants was ascertained. Field experiment was conducted at Krishi Vigyan Kendra, Research Farm, Janjgir – Champa (C.G.) during *kharif* season 2016. The increasing doses of fly ash with and without FYM show increasing content of heavy metals in soil and plant parts. The occurrence of heavy metals in paddy field soils was in a decreasing order of Cr > Pb > Co >Ni> Cd while, in plant parts order of Ni > Cr > Co >Pb> Cd. The highest concentration of heavy metals in this study was in the roots rather than in the parts of the rice plant that were above the ground (grain, leaf and stem), except for Cd, which was present in significant amounts in the leaves. However, these concentrations of heavy metals in the soil and rice plant parts were still below the maximum permissible limits.

Keywords: Fly ash, FYM, plant parts and heavy metals

Introduction
The ill effect of Industrialization all over the world is production of huge quantities of industrial waste that has arise challenges in front of administrator, environmentalist and scientists for its safe disposal and proper management is a major concern throughout the world. Presently about 118 Mt/year of coal FA is produced in India from more than 80 thermal power plants, which will be about 440 Mt/year by the end of 2030 (Ram *et al*., 2008) [15]. The production of vast quantity of FA will create dumping and management problems. The management of this huge amount of solid waste, at both regional and global level is a prime concern for the present and coming future (Ahmaruzzaman, 2010; Kishore *et al*., 2009) [1, 8], therefore agricultural utilization and waste area management techniques have emerged as prime utilization methods for solving fly ash problems. Field and green house experiments have proved that it can be used in growing agricultural crops (Gupta *et al*., 2002) [7].
Fly ash is an amorphous mixture of ferro-aluminosilicate minerals. Its physico-chemical characteristics depend on composition of parent coal, combustion conditions, the efficiency and type of emission control devices and the disposal methods used.
It contains small quantity of various essential plant major, secondary and micronutrients such as N, P, K, Ca, Mg, S, Fe, Mn, Zn, Cu, and B (Pandey *et al*., 2009) [13] and also due to availability of different pH fly ash, it will not only be used as a supplemental source of plant nutrients to correct the deficiency but also an amendment for improving acid, alkaline and degraded soils and thereby enhanced the physico-chemical properties of soils.
As it is known to contain quantities of heavy metals such as As, Cd, Cr, Ni, Mo, Pb, Se and Co. A major concern regarding FA augmented soil is linked with elevated level of heavy metal soil status and their accumulation through uptake by crops and associated risk in humans and animal health through its consumption. Therefore presence of toxic metals in FA may be serious concern for agriculture and there is a need to quantify these metals status in soil and plant through long-term application of fly ash.
Consistent accumulation of heavy metals due to continuous application of fly ash in paddy soils stresses the need for continuous monitoring and investigating several inter-related issues of our great concern. Keeping all these in mind, the present experiment has been designed to study the impact of varied levels of fly ash on toxic metal accumulation in different depths of soil and partitioning in different plant parts of rice (*Oryza sativa* L.).

Materials and Methods

A field experiment was carried out in Krishi Vigyan Kendra, Research Farm, Janjgir – Champa (C.G.) during *kharif* season 2016. The fly ash used in this study was collected from Madhya Bharat Paper Ltd. Village – Birgahni Champa, Dist. - Janjgir Champa, Chhattisgarh. The experiment consisted of eight treatments and each replicated thrice in a random block design. The treatments are Control, GRD (100:60:40), 75% GRD + 20 t ha^{-1} fly ash, 75% GRD + 40 t ha^{-1} fly ash, 75% GRD + 60 t ha^{-1} fly ash, 75% GRD + 20 t ha^{-1} fly ash + 5 t ha^{-1}FYM, 75% GRD + 40 t ha^{-1} fly ash + 5 t ha^{-1} FYM and 75% GRD + 60 t ha^{-1} fly ash + 5 t ha^{-1} FYM.

The rice cultivar Rajeshwari was selected as test crop and twenty five days old rice seedlings were transplanted in 20 x 15 cm spacing. The fly ash, fertilizer and the FYM were applied as per treatments one day ahead of transplanting. Soil samples for physical and chemical analysis were collected from the different layers viz. 0-15 and 15-30 cm after harvest of rice crop. The soil sample analyzed for different soil properties by following different standard procedure. The pH was measured with pH meter using 1:2.5 soil water suspensions (Black, 1965) [3]. The clear suspension extract obtained from pH was also utilized for EC measurement using conductivity bridge (Black, 1965) [3]. Soil texture was determined by International Pipette method as described by Piper (1966) [14]. The heavy metals (Ni, Co, Cr, Pb and Cd) (Lindsay and Norvell, 1978) [9] were analyzed by atomic absorption spectrophotometer.

Plant roots were extracted from the soil by washing in a container. Roots were washed again by deionized water and dried in an oven at 65^0C for 24 hours. Above ground parts of the plants were cut from the soil surface and dried in an oven at 65 ^{0}C for 24 hours. All parts were grinded and analyzed. The samples were acid-digested by using a digestion method devised for the analysis of heavy metals. Samples were analyzed by Atomic Absorption Spectrophotometer using respective holocathode lamps at desired wave lengths.

Table 1: The initial soil physico-chemical properties of experimental soil

Particulars	Analysis value	Soil type
Soil texture Sand (%) Silt (%) Clay (%)	 30 30 40	Clay loam
Soil reaction (pH)	7.5	
Electrical conductivity (dS m^{-1})	0.13	
Organic carbon (%)	0.44	
Available Cr (mg kg^{-1})	1.83	
Available Pb (mg kg^{-1})	1.14	
Available Ni (mg kg^{-1})	0.98	
Available Co (mg kg^{-1})	0.94	
Available Cd (mg kg^{-1})	0.22	

Results and Discussion

Characterization of soil, fly ash and FYM: The physico-chemical properties of soil, fly ash and FYM is given in table 1 & 2. The soil in was clay loam in texture, neutral in reaction, normal in soluble salts. Heavy metals present in soil are under the permissible limit. The heavy metals contents of soil followed the order Cr>Pb>Ni>Co>Cd whereas in fly ash, the order was Cr>Co>Ni>Pb>Cd.

Table 2: Physico-chemical properties of fly ash and FYM

Particulars	Fly ash	FYM
(0.02- 2 mm) Particle size distribution (0.002-0.02mm) (< 0.002 mm)	50 33 17	
Soil reaction (pH)	7.90	-
Electrical conductivity (dS m^{-1})	0.21	-
Organic carbon (%)	0.23	3.12
Total Cr (mg kg^{-1})	41.50	14.85
Total Co (mg kg^{-1})	29.20	17.32
Total Ni (mg kg^{-1})	31.80	13.00
Total Pb (mg kg^{-1})	16.52	10.21
Total Cd (mg kg^{-1})	13.50	1.60

Heavy metals (Ni, Co, Cr, Pb and Cd) contents in fly ash amended soil

The concentration of soil extractable Ni, Co, Cr, Pb and Cd in fly ash treated soils at various depths are given in table 3. The examination of table 3 and 4 denote that the concentrations of soil extractable Ni, Co, Cr, Pb and Cd in fly ash treated soils build up continuously as compared to control and 100% GRD. The average nickel status in surface and subsurface soil depths is shown in table 3. The effect of different treatments on the status of available Ni was found significant at both the depths. In surface soil, the application of different treatments was significantly increased the Ni status in fly ash applied up to 40 t ha^{-1}without FYM and treatment 75% GRD + 60 t ha^{-1} fly ash + 5 t ha^{-1}FYM when compare to 100% recommended dose of NPK. The application of fly ash @ 60 t ha^{-1}+ 75% GRD without FYM showed the highest (1.22 mg kg^{-1}) available Ni status, while the control showed the lowest (1.00 mg kg^{-1}).

In 15-30 cm soil depth, fly ash applied 20 and 40 t ha^{-1} fly ash with 75% GRD + 5 t ha^{-1} FYM and treatment 75% GRD + 60 t ha^{-1} fly ash are significantly increased the Ni status when compare to 100% recommended dose of NPK. The highest (0.84 mg kg^{-1}) Ni in treatment received is 75% GRD + 40 t ha^{-1} fly ash + 5 t ha^{-1} FYM.

Table 3: Effect of fly ash doses and in combinations with and without FYM on soil available nickel and cobalt content of different depth at harvest

	Available heavy metals (mg kg^{-1})			
	Ni		Co	
Treatment	0-15	15-30	0-15	15-30
T_1-Control	1.00	0.68	1.05	0.85
T_2- GRD (100:60:40)	1.09	0.70	1.11	0.88
T_3- 75% GRD + 20 t ha^{-1} fly ash	1.13	0.77	1.24	0.92
T_4- 75% GRD + 40 t ha^{-1} fly ash	1.19	0.78	1.28	0.94
T_5- 75% GRD + 60 t ha^{-1} fly ash	1.22	0.81	1.28	0.97
T_6- 75% GRD + 20 t ha^{-1} fly ash + 5 t ha^{-1} FYM	1.11	0.76	1.19	0.90
T_7- 75% GRD + 40 t ha^{-1} fly ash + 5 t ha^{-1} FYM	1.13	0.84	1.21	0.91
T_8-75% GRD + 60 t ha^{-1} fly ash + 5 t ha^{-1}FYM	1.16	0.80	1.25	0.92
SEm±	0.01	0.03	0.02	0.06
CD (P=0.05)	0.05	0.09	0.08	NS

The average cobalt status at both the soil depths is shown in table 3. The effect of different treatments on the status of soil available cobalt was found significant at surface and non significant at subsurface soil depth. At 0-15 soil depth, the application of different treatments was significantly increased the cobalt status with and without FYM when compare with control and 100% recommended dose of NPK. The application of fly ash @ 60 t ha^{-1}+75% GRD without FYM showed the highest in surface (1.28 mg kg^{-1}) and subsurface (0.97 mg kg^{-1}) soil depth available cobalt status while the control showed the lowest.

The soil available chromium status at different depths was found significant at surface soil and no significantly influence by application of fly ash with and without FYM when compared with control and 100% recommended dose of NPK (100:60:40) (Table 3). The available Cr status was the highest (2.59 mg kg^{-1}) in 0-15 cm and (1.97 mg kg^{-1}) in 15-30 cm soil depth with the application of 60 t ha^{-1} fly ash + 75% GRD without FYM.

Table 4: Effect of fly ash doses and in combinations with and without FYM on soil available chromium, lead and cadmium content of different depth at harvest

	Available heavy metals (mg kg^{-1})					
	Cr		Pb		Cd	
Treatment	0-15	15-30	0-15	15-30	0-15	15-30
T_1-Control	1.88	1.36	1.02	0.72	0.26	0.20
T_2- GRD (100:60:40)	1.95	1.47	1.08	0.74	0.29	0.22
T_3- 75% GRD + 20 t ha^{-1} fly ash	2.36	1.70	1.35	0.78	0.41	0.26
T_4- 75% GRD + 40 t ha^{-1} fly ash	2.53	1.82	1.33	0.84	0.44	0.28
T_5- 75% GRD + 60 t ha^{-1} fly ash	2.59	1.97	1.39	0.90	0.54	0.29
T_6- 75% GRD + 20 t ha^{-1} fly ash + 5 t ha^{-1} FYM	2.30	1.68	1.31	0.70	0.37	0.22
T_7- 75% GRD + 40 t ha^{-1} fly ash + 5 t ha^{-1} FYM	2.49	1.79	1.36	0.78	0.37	0.25
T_8-75% GRD + 60 t ha^{-1} fly ash + 5 t ha^{-1}FYM	2.51	1.84	1.48	0.86	0.41	0.24
SEm±	0.04	0.19	0.03	0.05	0.06	0.01
CD (P=0.05)	0.12	NS	0.09	NS	NS	NS

The average led status in surface and subsurface soil depths is shown in table 4. The effect of different treatments on the status of available Pb was found significant at surface soil depth but no change was observed in subsurface soil depth and there was a marginal increase over control and GRD. In surface soil, the application of different treatments was significantly increased the Pb status with and without FYM when compare with control and GRD (100:60:40). The application of fly ash @ 60 t ha^{-1}+ 75% GRD with FYM showed the highest (1.48 mg kg^{-1}) Pb status while the control showed the lowest (1.02 mg kg^{-1}).

Effect of different doses of fly ash integrated with and without on the status of available Cd was non significant at both the depths. The application of fly ash @ 60 t ha^{-1} +75% GRD without FYM showed the highest (0.54 and 0.29 mg kg^{-1}) in 0-15 and 15-30 cm soil depth respectively available Cd status while the control showed the lowest (Table 4).

The increasing doses of fly ash with and without FYM show increasing status of heavy metals content this might be due to fact that fly ash content of the toxic metals. The heavy metals status was higher at the surface than the subsurface depth. The occurrence of heavy metals in paddy field soils was in a decreasing order of Cr > Pb > Co >Ni> Cd at the surface and subsurface soil. Similar observation regarding to the metal content found by Gond *et al.* (2013) [6], Yeledhalli *et al.* (2008) [21] and Singh and Agrawal (2007) [17].

The distribution pattern of metal is governed by various soil physico-chemical factors like particle size fractions, pH, EC, organic matter, $CaCO_3$, cation exchange capacity, exchangeable cations etc. Datta *et al.* (2000) [5] studied the concentration of heavy metals declined with depth which may be due to lower permeability and vertical movement of the metals. The results also indicate that organic carbon plays a major role in mobility and transport of Cd, Ni, Cr and Pb in the soils. The immobilization of metals might have been due to adsorption and occultation on the surface by hydroxides and oxides in soils and tend to remain in the zone of incorporation. Similar observations have also been reported by Ciurl (2001) [4] and Appavu and Sree Ramulu (1990) [2].

Effect of fly ash on heavy metal accumulation in plant parts of rice

The effect of different doses fly ash integration with and without FYM on chromium, cobalt, nickel, and lead and cadmium concentration in different plant parts of rice presented in table 5 and 6.

The nickel concentration in different plant parts of rice influenced by graded dose of fly ash are non significantly increased in all parts (root, grain, leaf and stem) of the plant however marginal increased as compare to control and 100% GRD (Table 5). Fly ash applied @ 60 t ha^{-1} without FYM recorded the highest Ni content in root (14.73 mg kg^{-1}), leaf (13.12 mg kg^{-1}) and stem (9.14 mg kg^{-1}) however in grain highest(10.22 mg kg^{-1}) in application of 60 t ha^{-1} with FYM while, control showed the lowest.

The cobalt accumulation in different plant parts of rice influenced by different doses of fly ash with and without organic manure are shown in table 5. The result of cobalt content in leaf are significantly influenced by different fly ash treatments and all other plant parts i.e. root, grain and stem are non significant. In leaf Co content is significantly increased of all fly ash treatments with and without FYM as compare to control. Application of fly ash @ 60 t ha^{-1} without FYM is highest (5.17 mg kg^{-1}) and significantly increased Co content over the 100% recommended dose of NPK. Fly ash applied @ 60 t ha^{-1} without FYM recorded the highest Co content in root (5.60 mg kg^{-1}), grain (3.13 mg kg^{-1}) and stem (3.77 mg kg^{-1}) while, control was the lowest.

Table 5: Effect of fly ash doses and in combinations with and without FYM on nickel and cobalt content of different plant parts of paddy at harvest

	Ni content (mg kg^{-1})				Co content (mg kg^{-1})			
Treatment	**Root**	**Grain**	**Leaf**	**Stem**	**Root**	**Grain**	**Leaf**	**Stem**
T_1-Control	14.03	8.56	10.80	6.86	5.07	1.99	3.58	2.75
T_2- GRD (100:60:40)	13.90	9.44	11.77	8.24	4.97	2.73	4.16	3.23
T_3- 75% GRD + 20 t ha^{-1} fly ash	14.17	9.75	10.73	8.28	5.27	2.36	4.40	3.30
T_4- 75% GRD + 40 t ha^{-1} fly ash	14.23	9.80	12.08	9.03	5.30	2.84	4.50	3.67
T_5- 75% GRD + 60 t ha^{-1} fly ash	14.73	9.85	13.12	9.14	5.60	3.13	5.17	3.77
T_6- 75% GRD + 20 t ha^{-1} fly ash + 5 t ha^{-1} FYM	13.93	8.98	11.71	8.32	4.90	2.39	3.95	2.83
T_7- 75% GRD + 40 t ha^{-1} fly ash + 5 t ha^{-1} FYM	14.20	9.74	12.18	8.27	5.47	2.77	4.52	2.95
T_8-75% GRD + 60 t ha^{-1} fly ash + 5 t ha^{-1}FYM	14.53	10.22	12.48	8.36	5.57	2.85	4.58	3.73
SEm±	0.28	0.50	0.52	0.62	0.48	0.37	0.21	0.31
CD (P=0.05)	NS	NS	NS	NS	NS	NS	0.63	NS

The chromium concentration in grain and leaf are significantly increased and non significant influence in root and stem part of rice by different fly ash treatments (Table 6). In grain all the treatments of fly ash with and without organic fertilizer significantly increase over control but statistically at par over 100 % recommended dose of NPK. Among the treatments 75% GRD + 40 t ha^{-1} fly ash recorded the highest (5.17 mg kg^{-1}) content and control showed the lowest (3.99 mg kg^{-1}). Fly ash applied @ 40 and 60 t ha^{-1} without FYM significantly influenced chromium content in leaf over 100 % recommended dose of NPK and all other treatments similar to 100% GRD. Among the treatments 75% GRD + 60 t ha^{-1} fly ash recorded the highest (8.10 mg kg^{-1}) content and control showed the lowest (7.37 mg kg^{-1}).

The lead accumulation in grain is significantly influenced by different fly ash treatments and all other plant parts i.e. root, leaf and stem are non significant (Table 6). The result of cobalt content in grain are significantly influenced by different fly ash treatments over control. Application of fly ash 40 and 60 t ha^{-1} without FYM and 60 t ha^{-1} fly ash with FYM recorded significantly increased over 100 % recommended dose of NPK. Application of 75% GRD + 60 t ha^{-1} fly ash recorded the highest in root (3.84 mg kg^{-1}), grain (3.23 mg kg^{-1}) and stem (3.17 mg kg^{-1}) Pb content and control showed the lowest.

Table 6: Effect of fly ash doses and in combinations with and without FYM on chromium, lead and cadmium content of different plant parts of paddy at harvest

	Cr content (mg kg^{-1})				Pb content (mg kg^{-1})				Cd content (mg kg^{-1})			
Treatment	**Root**	**Grain**	**Leaf**	**Stem**	**Root**	**Grain**	**Leaf**	**Stem**	**Root**	**Grain**	**Leaf**	**Stem**
T_1-Control	6.90	3.99	7.37	4.96	3.15	2.00	2.73	1.97	2.16	1.79	2.45	1.81
T_2-GRD (100:60:40)	7.20	4.52	7.56	5.63	3.47	2.03	2.70	2.27	2.73	1.88	2.72	2.00
T_3- 75% GRD + 20 t ha^{-1} fly ash	7.21	4.62	7.87	5.65	3.67	2.04	2.73	2.17	2.78	1.92	2.78	2.01
T_4- 75% GRD + 40 t ha^{-1} fly ash	7.38	5.17	8.02	5.76	3.47	3.12	2.83	2.23	2.82	1.96	3.12	2.01
T_5- 75% GRD + 60 t ha^{-1} fly ash	7.66	5.11	8.10	6.02	3.84	3.23	3.10	3.17	1.88	2.00	3.24	2.12
T_6- 75% GRD + 20 t ha^{-1} fly ash + 5 t ha^{-1} FYM	6.86	4.61	7.43	5.66	3.37	2.06	2.70	2.23	2.76	1.87	2.74	1.99
T_7- 75% GRD + 40 t ha^{-1} fly ash + 5 t ha^{-1} FYM	6.99	4.63	7.95	5.99	3.60	2.10	3.15	2.37	2.92	1.91	2.80	2.12
T_8-75% GRD + 60 t ha^{-1} fly ash + 5 t ha^{-1}FYM	7.67	4.72	7.97	6.13	3.37	3.16	3.03	2.27	2.94	1.96	2.81	2.11
SEm±	0.30	0.19	0.15	0.33	0.15	0.23	0.19	0.24	0.45	0.08	0.10	0.06
CD (P=0.05)	NS	0.60	0.45	NS	NS	0.70	NS	NS	NS	NS	0.30	NS

The cadmium concentration in different plant parts i.e. root, grain and stem of rice was non significantly influenced but in leaf are significantly increased by application of fly ash integrated either with or without FYM (Table 6). In leaf application of 40 and 60 t ha^{-1} fly ash without FYM are significantly increased cadmium content as compare to 100% recommended dose of NPK. Cadmium concentration is recorded in maximum in leaf when fly ash applied without organic fertilizer and highest (3.24 mg kg^{-1}) concentration is found in application of 75% GRD + 60 t ha^{-1} fly ash.

The distribution pattern of heavy metals in different plant parts in a ranking order of Ni > Cr > Co >Pb> Cd. Among all the five (Pb, Cd, Cr, Ni and Co) assessed heavy metals Ni showed more accumulation in different plant parts.

Increasing doses of fly ash with and without FYM found increased concentration of heavy metals due to the high content of these trace metals in fly ash. Metal accumulation highest in different plant parts of rice when application of fly ash with chemical fertilizer and without FYM. The concentration of heavy metal elements in plant depends upon the content of those elements in the material used (Soil/ fly ash /FYM), the ratio of ash/ FYM application, the soil properties and the type of plant.

Fly ash used in the study contain higher amounts of heavy metals but the concentration of heavy metals in fly ash amended soil and the rice produced was within the maximum permissible limit reported by WHO after the application of fly ash highest dose (60 t ha^{-1}).

Mo *et al*. (2001) [11] reported that most of the heavy metals absorbed by the paddy plant accumulated in the roots, where there was a high metabolic rate and that the accumulation in the stem and leaves was low. The accumulation and distribution of heavy metals in the upper parts of a plant are determined by several factors, including anatomical, biochemical and physiological factors.

Singh *et al*. (2004) [16] also reported that the higher accumulation of heavy metals in roots may be ascribed to formation of complex between heavy metals and sulphydryl groups that results less transport of heavy metals to shoots.

The metal contents, physico-chemical properties of soil, organic substances and other elements in the rhizosphere affected the bioavailability and plant uptake of heavy metals from the soils. They also emphasized that plant roots and soil microbes and their interaction could improve the metal bioavailability in rhizosphere through secretion of proton, organic acids, phytochelatins (PCs), amino acid and enzymes. The distribution of heavy metals in different plant parts was also supported by the results of Maiti and Nandhini (2006) [10], Pandey *et al*. (2012) [12] and Tripathi *et al*. (2008) [15].

Conclusion

This study determined the accumulation of heavy metals in paddy soil and rice plant parts which were collected simultaneously at harvest period. The results revealed that the metal concentrations in the different soil depth that were amended by fly ash with and without FYM were below the maximum permissible limits. The findings also revealed that most of the heavy metals in the study accumulated mainly in the roots of the paddy plants except for Cd, which was retained in the leaves. Meanwhile, the metal concentrations in soil and the plant parts, including the root, grain, leaf and stem were less than the maximum permissible limits specified by WHO guidelines. The concentration of heavy metals in soil and plant parts increase with increasing doses of fly ash with and without FYM. However, FA dose optimization need extensive field trials and will vary based on soil type and source of fly ash. Besides, studies should need to examine the long term impact of FA application on heavy metals status.

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