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Effect of Fly Ash Applications on Heavy Metal Contents of Soil and Corn Plant (*Zea mays* L.)

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Abstract

In this study, the chemical composition of fly ash released from 18 Mart Çan Coal-Fired Power Plant was characterized and its effects on heavy metal contents of soil and corn plant were investigated. Six different fly-ash doses (0, 2, 4, 8, 16 and 24 % w/w) were mixed with soil in pots and corn seeds were sown. Two months later, total element concentrations of soil, roots and plant stems were determined. Although significant changes were not observed in aluminum (Al) and lead (Pb) contents with fly ash treatments, significant increases were observed in boron (B), sodium (Na), sulphur (S) and molybdenum (Mo) at the level of p< 0.01 for soil samples. Soil Calcium (Ca) and Iron (Fe) were changed at the level of p< 0.05. While Phosphorus (P), Chrome (Cr), Manganese (Mn), Nickel (Ni), Zinc (Zn), Magnesium (Mg), Potassium (K), Cadmium (Cd) and Lead (Pb) contents of corn roots and stems did not change significantly, there were significant increases in B, Na, Mg, S, Cupper (Cu), Cd, Mo, B, S , Mo (p < 0.01) and Mn (P < 0.05). This result is important in terms of heavy metal pathways and therefore food chains.

Keywords: Fly ash; Heavy metal; Soil; Corn

Uçucu Kül Uygulamalarının Toprak ve Mısır Bitkisinin Ağır Metal İçeriklerine Etkisi

Özet

Bu çalışmada, 18 Mart Çan Termik Santrali uçucu külünün kimyasal bileşimi karakterize edilmiş ve uçucu kül uygulamalarının toprak ve mısır bitkisinde ağır metal içerikleri üzerine etkileri araştırılmıştır. Altı farklı uçucu kül dozu (Ağırlık esasına göre % 0, 2, 4, 8, 16 ve 24) karıştırılan saksılara mısır tohumları ekilmiştir. İki ay sonra toprakta, bitki köklerinde ve bitki saplarında toplam element konsantrasyonları belirlenmiştir. Uçucu kül uygulamaları ile Alüminyum (Al) ve Kurşun (Pb) konsantrasyonlarında önemli değişiklikler olmamasına rağmen, önemli artışlar; Bor (B), Sodyum (Na), Kükürt (S) ve Molibden (Mo) elementlerinde (p < 0.01) seviyesinde, Kalsiyum (Ca) ve Demir (Fe) elementlerinde (p < 0.05) seviyesinde gözlemlenmiştir. Mısır bitkisinin kök ve gövdesinde; Fosfor (P), Krom (Cr), Mangan (Mn), Nikel (Ni), Çinko (Zn), Magnezyum (Mg), Potasyum (K), Kadmiyum (Cd) ve Pb artışları önemsiz olurken; B, Na, Mg, S, Bakır (Cu), Cd, Mo, B, S ve Mo elementlerinde (P <0.01) düzeyinde, Mn elementinde ise (P < 0.05) derecesinde istatistiki olarak önemli değişimler görülmüştür. Sonuçlar; ağır metal taşınım yolları ve besin zincirleri bakımından önemlidir.

Anahtar kelimeler: Uçucu kül, Ağır metal, Toprak, Mısır

Introduction

Coal-fired thermal power plants are one of the most common means of energy production in the world. Fly ash particles ranging in size from 0.01 to 100 μ m are the ultimate combustion by-product of coal (Davison et al., 1974). The phases,

mineral and chemical compositions and possible uses of fly ash particles were studied by several researchers (Karayiğit et al., 2000; Vilches et al., 2005; Inada et al., 2005; Stanislav and Rosa, 2005; Arvelakis and Frandsen, 2005; Mishra and Das, 2010; Garg and Pundir, 2012; Bhattacharjee et al., 2013; Kaewmanee et al., 2013). Fly ash is a heterogeneous material and SiO₂, Al₂O₃, Fe₂O₃ and occasionally CaO are the main chemical components present in fly ashes. The main phases encountered are glass phase, together with guartz, mullite and the iron oxides hematite and magnetite. Fly ash is generally stored at coal power plants or placed in landfills in the USA. About 43% is recycled, often used to supplement Portland cement in concrete production. It is collected through electrostatic precipitators and is potentially hazardous industrial waste due to toxicity associated with the constituent trace elements concentration (Adriano et al., 1980). Fly ash is an also a part of solid waste, which creates problem not only for human being but also for environment. It is known to contain significant quantities of heavy metals such as arsenic, lead and selenium, which can cause cancer and neurological problems.

Although it contains potentially harmful trace elements such as As, Cd, Mo, Pb, Se and Sb, it also contains mineral constituents such as Cu, Fe, Zn, Mn and Mo and hence, can act as a secondary source of fertilizer (Pandey et al., 2009). Fly ash, with its abundant availability and remarkable ameliorative and nutritive properties, warrants an eco-friendly approach for waste materials as a soil amendment. Fly ash may be used in plant growth and soil reclamation but scientific research is certainly necessary to model the concentration-uptake-dose-response functions between the amended medium and plants (Jala and Goyal, 2006; Spark and Swift 2008; Pandey et al., 2009; Ansari et al., 2011; Ritchey et al., 2012). Some studies on different plants and certain tree species have focused on acidic soils with low level of fly ash application (Mishra and Shukla, 1986).

In present study, phase, mineral and chemical properties of fly ashes from 18 Mart Çan Coal-Fired Power Plant (Turkey) were investigated and the effects of fly ash treatments on heavy metal contents of soils and plants were evaluated.

Materials and Methods

Six different fly-ash doses (0, 2, 4, 8, 16 and 24 % w/w) were mixed with experimental soil in pots. Three corn seeds (DKC 6842) were sown in each pot and then it was thinned to one after the emergence. Plants were grown in pots under greenhouse conditions. Experiments were conducted in randomized block design with 4 replications for each fly ash dose. Experiments lasted for 60 days.

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°C

for 24 hours. Above ground parts of the plants were cut from the soil surface and dried in an oven at 65 °C for 24 hours. All parts were grinded and analyzed. The samples were acid-digested by using a special two-step digestion method devised for the analysis of trace elements in coal and combustion wastes by Inductively Coupled Plasma Mass Spectrometry (ICP-MS). This is based on initially HNO₃ extraction in closed PFA reactors under 90 °C for 6 hours to extract the volatile metals, followed by the microwave digestion of the solid residue (isolated with centrifugation) with HF:HNO3:HClO4. Finally, the resultant extract was dried out and the soluble residue re-solved by using HNO₃. Heavy elements were analyzed in acid digestions with an ICP-MS by using the analytical conditions specified in literature (Tait and Ault, 1992; Querol et al., 1995). Lastly, C and N contents were determined in accordance with Kirsten (1983). Other major, minor and trace element concentrations of fly ash, soil and corn plants were determined according to Table 1.

Experimental results were statistically analyzed by MINITAB 16.0 statistical analysis software. Differences among the means were tested by Least Significant Difference (*LSD*) test at 5% significance level.

Results and Discussion

The element composition of fly ash and soil are provided in Table 1. As it can clearly be seen from the table, the element contents in fly ash particles and soil were mainly composed of atmophile (C and N), lithophile (B, Na, Mg, Al, P, K, Ca, Cr and Mo), chalcophile (S, Cu, Zn, Se, Cd, and Pb) and siderophile (Mn, Fe and Ni) elements. Lithophile elements in fly ash particles are more dominant than siderophile elements. The concentration of elements ordered from the highest to lowest as Ca, Fe, Mg, Na and S for fly ash. K, B, P and Mn were at substantial levels in fly ash. There were significant differences between element content of fly ash and soil. For instance, fly ash has higher concentrations of Na, Mg, Al, S, Ca, Fe, Cu, Zn, Se and Mo, whereas soil has greater contents of P, K, Mn, Cr, Ni and Pb. The amount of nitrogen and carbon in the soil were higher than fly ash.

Effects of fly ashes on soil and corn plants are presented in Tables 2, 3 and 4. Table 2 shows that fly ash treatments increased B, Na, Mg, P, S, K, Ca, Al, Cr, Mn, Fe, Cu, Zn, Se and Mo contents of soil. B (P < 0.01), Na (P < 0.01), S (P < 0.01), Ca (P < 0.05), Fe (P < 0.05) and Mo (P < 0.01) contents significantly increased with increasing fly ash doses. Increasing element contents are directly related to fly ash treatment doses.

Properties	Soil	Methods of Analyze	Fly ash	Methods of Analyze
Texture	SCL	Bouyoucous (1951)		
рН	7.47	Soil:Water (1:2.5 w/w),	7.72	Fly ash:Water (1:10 w/w),
	7.47	Richards (1954)	1.12	Richards (1954)
Salinity (dS/m)	1.64	Soil:Water (1:2.5 w/w),	4.28	Fly ash:Water (1:10 w/w),
		Richards (1954)		Richards (1954)
CaCO₃ (g/kg)	28.50	Scheibler calcimeter,	14.30	Scheibler calcimeter,
		Allison and Modie (1965)	11.50	Allison and Modie (1965)
N (g/kg)	3.10	Leco C-N Analyzer dry combustion Kirsten	0.34	Leco C-N Analyzer dry
(6/	0.120	(1983)		combustion, Kirsten (1983)
C (g/kg)	29.80	Leco C-N Analyzer dry combustion Kirsten	17.60	Leco C-N Analyzer dry
	23.00	(1983)	17.00	combustion, Kirsten (1983)
Avail. P (µg/g)	74.80	After 0.5M NaHCO3 extraction Olsen et	53.34	0.5M NaHCO ₃ extraction, Olsen
(1-8/8/		al. (1954)		et al. (1954)
Elements –		(Total mg/g)*		
	Soil		Fly ash	
Potassium (K)	6.62		3.51	
Calcium (Ca)	16.36		23.39	
Magnesium (Mg)	3.33		2.29	
Sodium (Na)	1.16		9.81	
Sulphur (S)	1.20		8.87	
Iron (Fe)	15.85		23.13	
Elements		(Total μg/g)*		
Phosphorus (P)	794.50		562.19	
Manganese (Mn)	689.30		441.90	
Boron (B)	26.77		1079.00	
Copper (Cu)	38.81		61.11	
Zinc (Zn)	77.72		93.93	
Aluminum (Al)	56.12		78.61	
Chrome (Cr)	30.57		18.40	
Nickel (Ni)	26.70		17.67	
Lead (Pb)	22.15		18.98	
Selenium (Se)	3.95		9.57	
Molybden. (Mo)	0.88		9.31	
Cadmium (Cd)	0.73		0.73	
Cobalt (Co)	0.00		0.00	

Table 1. Some properties and element concentrations of the fly ash and soil

*: Total elements were determined with ICP-MS instrument after wet digestion with an HF:HNO₃:HClO₄ on PFA reactors under 90 °C according to Tait and Ault, 1992; Querol et al., 1995.

The highest increase was observed in pots containing 24% fly ash. With fly ash treatments, the largest increase was seen in B and it was followed by S (Krauskopf, 1972). Basu et al., (2009) reported that fly ash boron content levels were between 10-618 mg kg⁻¹ in countries such as India, China, USA, Germany, UK, Australia, Canada, France, Denmark, Italy and Netherlands.

Table 3 shows that fly ash treatments also increased B, Na, Mg, P, S, Ca, Al, Fe, Cu, Se and Mo contents of plant roots. B (P < 0.01), Na (P < 0.01), Mg (P < 0.01), S (P < 0.01), Cu (P < 0.01), Cd (P < 0.01) and Mo (P < 0.01) contents significantly increased and increasing element contents were directly related to fly ash treatment doses. B, Na, P, S, Ca, Mn, Cu, Zn, Cd and Mo also increased with fly ash treatments on above-ground parts of plant.

But the contents of B (P < 0.01), S (P < 0.01), Mn (P < 0.05) and Mo (P < 0.01) significantly increased at the same part of plant (Table 4). Increasing element contents were directly related to fly ash treatment doses. Corn plants contained much more B when fly ash was applied to soil (Müftüoğlu et al., 2012; Müftüoğlu et al., 2013) due to excess boron content of raw materials. Additionally fly ash that was taken from 18 Mart Çan Coal-Fired Power Plant had up to 1000 µg g⁻¹ boron (Table 1).

Karayiğit et. al. (2000) reported that boron content of coal samples were 1.4-244 μ g g⁻¹ (Çayırhan, Seyitömer, Tunçbilek, Orhaneli, Soma, Yatağan, Yeniköy, Elbistan, Kangal and Çatalağzı). Especially coals from Çayırhan, Seyitömer, Tunçbilek (A3), Soma (B1-4 and B5-6) lignite boron contents were >100 μ g g⁻¹.

Elements	Fly ash doses (%)						
(µg/g)	0	2	4	8	16	24	—LSD
К	6622	8148	7186	6940	7475	7395	ns
Ca	16359 ab	17471 a	13701 b	14139 b	14854 ab	16917 a	p < 0.05
Mg	3325	3441	3296	3259	3324	3388	ns
Na	1155 d	1503 c	1390 cd	1692 c	2107 b	3095 a	p < 0.01
Р	795	822	755	803	776	723	ns
S	1202 d	1396 d	1402 d	1796 c	2193 b	2771 a	p < 0.01
Fe	15847 b	17575 ab	17563 ab	17769 ab	18493 a	19788 a	p < 0.05
Mn	689	717	717	790	672	673	ns
Zn	77.72	76.86	73.06	76.10	81.95	79.63	ns
Cu	38.81	34.29	36.16	40.23	41.03	40.20	ns
В	26.77 e	47.88 d	58.33 d	80.41 c	123.47 b	222.20 a	p < 0.01
Мо	2.35 d	2.36 d	2.80 cd	3.51 c	5.78 b	15.23 a	p < 0.01
Ni	26.70	29.42	23.60	22.92	24.67	27.51	ns
Se	3.95	6.30	3.71	4.15	5.88	5.88	ns
Al	56	61	60	62	67	66	ns
Cd	0.734	0.606	0.534	0.530	0.623	0.567	ns
Cr	30.57	33.91	29.77	25.75	35.75	42.90	ns
Pb	22.15	22.11	21.92	21.46	21.85	22.92	ns

Table 2. The effect of fly ash on heavy metal content of the soil

LSD: Least Significant Difference, ns: Not Significant

Rows labeled with the different letters are significantly different at p < 0.05 or p < 0.01 according to LSD test.

Elements	Fly ash doses (%)						
(µg/g)	0	2	4	8	16	24	—LSD
К	34616	32871	32719	34181	35896	29823	ns
Ca	6355 b	5555 b	5484 b	6392 b	5836 b	12950 a	<i>p</i> < 0.01
Mg	2395 b	2269 b	2236 b	2300 b	2262 b	2897 a	<i>p</i> < 0.01
Na	7493 bc	5711 cd	4988 d	6488 bcd	8022 b	14474 a	<i>p</i> < 0.01
Р	2562 b	2736 b	2897 b	2469 b	2610 b	2255 a	ns
S	3869 d	4240 cd	4546 c	4758 c	5658 b	7155 a	<i>p</i> < 0.01
Fe	2723	2999	3255	2584	2210	3308	ns
Mn	97	97	104	79	57	94	ns
Zn	58.02	47.66	44.56	52.56	50.20	56.81	ns
Cu	10.72 b	10.47 b	11.06 b	10.78 b	10.02 b	16.01 a	<i>p</i> < 0.01
В	4.12 b	17.96 b	7.91 b	22.67 b	69.08 b	261.10 a	<i>p</i> < 0.01
Мо	0.88 e	1.23 d	1.36 d	1.61 c	1.98 b	2.74 a	<i>p</i> < 0.01
Ni	13.52	16.94	11.18	13.97	9.62	12.40	ns
Se	0.24	1.11	0.12	0.69	0.51	1.45	ns
Al	497	620	608	552	512	599	ns
Cd	0.227 bc	0.180 c	0.189 c	0.258 bc	0.292 b	0.415 a	<i>p</i> < 0.01
Cr	5.11	5.86	5.38	4.70	4.06	4.92	ns
Pb	1.09	1.45	1.77	0.67	0.60	1.62	ns

LSD: Least Significant Difference, ns: Not Significant

Rows labeled with the different letters are significantly different at p < 0.01 according to LSD test.

Elements		Fly ash doses (%)					
(µg/g)	0	2	4	8	16	24	- LSD
К	40501	39052	38800	40918	40199	44783	ns
Ca	3333	2904	2915	2996	3566	3743	ns
Mg	2002	1885	1894	1885	2030	1942	ns
Na	754	402	382	468	446	1058	ns
Р	3228	3940	4273	3242	3927	2520	ns
S	1280 c	1246 c	1294 c	1386 c	1721 b	2480 a	p < 0.01
Fe	197	123	101	104	196	127	ns
Mn	41 bc	41 bc	45 ab	43 ab	52 a	34 c	p < 0.05
Zn	31.00	29.71	25.00	33.34	40.05	24.99	ns
Cu	4.95	7.59	4.20	10.22	13.90	5.95	ns
В	34.57 d	100.20 cd	128.20 cd	268.50 c	696.50 b	1230 a	p < 0.01
Мо	1.94 c	1.97 c	2.15 c	2.56 c	4.02 b	8.45 a	p < 0.01
Ni	13.19	4.26	6.24	3.33	6.04	6.23	ns
Se	0.96	0.25	0.71	0.85	1.42	1.61	ns
Al	125	83	122	111	139	129	ns
Cd	0.052	0.042	0.089	0.068	0.047	0.052	ns
Cr	4.46	2.19	2.00	1.65	2.81	2.28	ns
Pb	0.00	0.00	0.00	0.15	0.58	0.00	ns

Table 4. The effect of fly ash on heavy metal content of above-ground parts of the plant

LSD: Least Significant Difference, ns: Not Significant

Rows labeled with the different letters are significantly different at p < 0.05 or p < 0.01 according to LSD test.

Yaman (1992) reported that compounds that form mineral matters depend on both geological environment as well as coals age therefore each coal has different element content and distributions. Additionally the same researcher reported that XRD analysis results of lignite samples from 18 Mart Çan Power Plant showed that it had Quarts, Tridymite, Hematite, Magnetite, Goethite, Rozenite, Kaolinite, Ca-SO4-hydrate, Pyrite; Fly ash samples had; Quarts, Hematite, Anhydride, Anorthite, Mullite and Gehlenite.

Gluskoter (1977) reported that tourmaline [Na(Mg,Fe)₃Al₆(BO₃)₃(Si₆O₁₈)(OH)₄] mineral had boron in form of BO₃. In this case, lignite coals that were used in 18 Mart Çan Power Plant contains neither tourmaline nor similar boron contained minerals are in conflict with the ashes with high boron content. The reason of high boron level is SO₂ production during combustion which is undesirable. It should be also considered that powdered CaCO₃ that is sprayed into the fluidized bed boiler during burning may contain boron and therefore its boron content needs to be determined at first.

Some researchers reported that fly ash had also negative effects on boron content of plant (Krauskopf, 1972) and plant growth (Adriano et al., 1980; Spark and Swift 2008; Pandey et al., 2009; Ansari et al., 2011; Ritchey et al., 2012; Bozyel, 2011).

Conclusion

It was the first time in present study that chemical composition of thermal power plant fly ash was investigated by Inductively Coupled Plasma Mass Spectrometry (ICP-MS). The lithophile elements (B, Na, Mg, Al, P, K, Ca, Cr and Mo) were more dominant than siderophile elements (Mn, Fe and Ni) in fly ash. Fly ash and soil show significant differences with regard to element contents. While fly as had higher Na, Mg, Al, Si, Ca, Fe, Cu, Zn, Se and Mo contents, soil had higher P, K, Mn, Cr, Ni, and Pb contents. Fly ash treatments significantly increased soil Fe (P < 0.05) and Mo (P < 0.01), corn root Cu (P < 0.01), Cd (P < 0.01) and Mo (P < 0.01) and plant stem Mn (P < 0.05) and Mo (P < 0.01) contents.

Element concentrations increased with increasing fly ash doses. Even it was though that application of fly ash may improve some soil characteristics, further research is recommended for better understanding of possible negative impacts of such treatments.

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