Influence of coal fly ash application on trace element mobility and distribution in soil, plant and leachate

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Abstract:
A greenhouse study was conducted to assess growth and elemental uptake by canola (*Brassica napus*) on acidic soils amended with four chemically different unweathered fly ashes. The ashes were incorporated into the top 10 cm of 1 m long intact soil cores at the rates of 0, 12, 36 and 108 Mg ha^{-1}. The objective of this study was to characterise growth responses and distribution of B, Mo and Se in leachate, soil and plant. We observed increases in the soil pH and that of the leachate within 5 months of the fly ash addition. Concentrations of Mo and Se increased significantly in the stem, while that of B was slightly increased, with 108 Mg ha^{-1} fly ash application. Compared with the untreated control, acidic fly ashes increased above ground biomass produced by canola by up to 25 %, but biomass was unchanged or reduced with the alkaline ashes. In general, concentrations of B, Mo and Se in canola seed increased with higher rates of fly ash. While concentrations of Mo and Se in the soil were only slightly increased, that of B was elevated significantly in the top 0-10 cm of the soil with the application of alkaline fly ashes. The concentrations of the trace elements in the leachate were generally below detection limit. Accumulation of the three metals in soil and plant were all below phytotoxic levels. The results of this study suggest that fly ash has the potential to increase plant growth without any detrimental effects on soil or groundwater. This is a limited study and there is a need for a continuous monitoring of trace elements in the soil-plant and water system under field conditions.

Introduction

Fly ashes can have beneficial effect on plant growth and yields when applied to soils because of their chemical, physical, mineralogical characteristics and nutrient content. However, a major concern is the presence of relatively high concentrations of trace elements such as As, Cd, Cr, B, Ni, Mo and Se, present in many ashes, which when added to the soil may cause toxicity and environmental hazard.[1] Although this may not be a major concern for the vast majority of Australian fly ashes that generally contain relatively lower concentrations of most of the trace elements.[2] Some of these elements are, however essential for plant growth and are required in trace amounts, but becomes toxic when available in excess amounts. An earlier study by Aitken and Bell[3] identified B as a main element of concern to plant growth and yield, because of its relatively high amounts and solubility in some of the Australian fly ashes. Many overseas studies also suggested that B toxicity is a main limitation in using fly ash for the land application.[4] In contrast to B, elements such as Mo and Se are considered to be essentially non-toxic to plants at the levels expected to result from fly ash amendments at common agronomic rates, but they have high tendency to bioaccumulate over time and become toxic in animals.[5] Therefore, any agronomic/environmental assessment of fly ash needs to consider both short- and long-term dynamics of these elements in the soil–plant system. Furthermore, Mo and Se become more soluble and easily available to the plant as pH increases following fly ash application to acidic soils.[4]

However, the amount of trace element taken up by plants from fly ash does also depends on plant species. While there are no definite distinctions amongst plant groups in terms of bioaccumulation of Mo, legumes appear to accumulate B, grasses Se[6] and brassica species B and Se.[7] In a pot experiment, Furr et al.[8] found that fly ash increased B, Mo, Se concentrations in the edible parts of beans, cabbage, millet and carrots. Later, Tolle et al.,[9] reported that lucerne (*Medicago sativa*) showed toxic accumulation of only B when grown on acidic soil treated with fly ash at between 0 and 700 Mg ha^{-1}.

Leaching of trace element in to groundwater is an important issue in the use of fly ash on agricultural lands. Several studies [10, 11] have examined the elemental leaching from fly ashes however, only a few studies have addressed the potential for trace elements to leach from the incorporation layer into subsoil horizons with fly ash application.[12, 13, 14]

Most of these previous plant growth studies used sandy soils and extremely high rates of fly ash often under non-leaching conditions. We are not certain how the responses in the elemental uptake, growth and yield of plant and leaching of trace elements are affected when fly ash is applied to medium or heavy textured soil at low to moderately high rates. In this study we used large intact soil cores as mimics of field soils to quantify growth and yield by canola, and concentrations of selected trace elements in the crop, soil and leachate, when four chemically different fly ashes were applied to the soil. The specific objectives were to (1) characterise distribution of Se, Mo and B in soil, plant and leachate, (2) quantify yield and seed trace element content for canola, and (3) determine basic chemical properties of the leachate.

Materials and methods

Soils and fly ashes used:

We used two representative agricultural soils from southern tablelands of New South Wales, Australia. Intact soil columns (100 cm deep and 15 cm internal diameter) were collected from two sites at Belangalo and kangaloon using ProLine hydraulic corers mounted on a tractor A set of bulk samples were collected from the top 25 cm of each soil and was air dried for chemical and physical analysis.

Treatments:

Each of the four ashes was mixed into top 10 cm of the soil at rates of 0, 12, 36 or 108 Mg ha^{-1} to set of cores for each of the two soil types. Each ash rate had three replicates on each soil. Eight seeds of canola (*Brassica napus*, cv. Surpass 603CL) were planted at depth of 20 mm in each pot. After emergence the seedling later were thinned to 4 per pot one week after emergence and latter to 2 plants/core at start of flowering. The cores were sealed at the base, but supplied with a drainage outlet that allowed leachate to be collected into bottles, which was done periodically.

Measurements:

Plant growth and yield: Canola shoots and seeds were harvested at maturity and their dry weights obtained after drying in a forced air oven at 60 ^{o}C for three days before being weighed. Dried plant samples were ground in a stainless steel grinder and sub-sample was used for chemical analysis.

Soil chemical characteristics:

After plant harvest, composite moist soil samples of 250 g were taken from the Belangalo soil only at various depths (0-10, 10-20, 20-40, 40-60-, 60-90 cm) from each soil core for chemical analysis. Soil cores from Kangaloon were replanted with beans to study the residual effects of fly ash application. The pH and EC for the soil were measured in 1:5 soil/water (w/v) suspensions. Concentrations of the trace and macro elements in all leachate were determined by inductively coupled plasma mass spectroscopy (ICP-MS). Prior to analysis, all soil, plant and seed samples were digested according to USEPA 5030b method. Extractable B determination in soil samples were carried out using the hot 0.01 M $CaCl_2$ extraction technique.[15] Calcium, Mg, K, and P were determined by ICP-OES and all other elements by ICP-MS.

Statistical Analysis
General linear models (GLMs) were used to determine significant differences within the data, using a probability level of $P<0.05$ in all cases.

Table 1. Selected chemical characteristics of the original (untreated) soils.

	Belangalo Soil (Brown Kandosol$^{\beta}$)	Kangaloon Soil (Red Ferrosol$^{\beta}$)
pH (1:5 soil: water)	5.02	5.41
EC (1:5 soil: water)-dSm^{-1}	0.072	0.222
CEC-$cmol^{(+)}kg^{-1}$	3.98	15.78
Na $cmol^{(+)}kg^{-1}$	0.05	0.65
Ca $cmol^{(+)}kg^{-1}$	1.69	10.23
Mg $cmol^{(+)}kg^{-1}$	0.85	4.34
K $cmol^{(+)}kg^{-1}$	0.20	0.37
P_{α}	10	8
SO_4-S	14	24
Organic matter-%	3.11	4.94
HWSB¶	0.72	1.65
Total B	46	98
Se#	<0.1	0.6
Mo#	0.66	0.41

#- Concentrated HNO_3 extractable (1:5 soil: Solution); ¶ Hot 0.01 M $CaCl_2$ extractable B; α- Bray 1 extractable P; β-According to Australian soil Taxonomy

3. RESULTS AND DISCUSSION

3.1. Chemical and Physical Properties of soils and fly ashes

Soils used in this study differed in their chemical, physical and nutritional properties (Table 1). Belangalo soil (sandy clay loam) had relatively low pH, CEC, electrical conductivity (EC), Hot water extractable B ($_{HWB}$) and other micro nutrients compared with Kangaloon soil (clay loam). Fly ashes also showed considerable variation in their chemical properties (Table 2). For example, the $pH_{(H2O)}$ ranged from 3.28 to 10.77 with Alkaline FA-I being the most alkaline and Acidic FA-I the most acidic. The electrical conductivity (EC) ranged from 0.14 to 19.06 dSM^{-1}, with Alkaline FA-II had the highest EC.

Table 2. Selected chemical characteristics of the fly ashes.

Fly Ash	pH^{α}	EC^{β} dSm^{-1}	HNO_3 soluble$^{\chi}$						
			Ca %	Mg %	P %	Mo $mgkg^{-1}$	Se mg kg^{-1}	B	B_{HW}
								mg kg^{-1}	
Alkaline FA-I	10.7	0.54	0.26	0.05	0.021	8.9	3.7	65	6.7
Acidic FA-I	3.2	1.10	0.49	0.16	0.338	33.9	9.6	12	3.1
Acidic FA-II	3.9	0.14	0.03	0.01	0.013	5.4	0.9	18	3.2
Alkaline FA-II	9.0	19.06	9.28	8.94	0.02	4.5	11.2	126	6.8

α-1:5 soil: water (w/v); β-electrical conductivity (EC) 1: 5 soil: water (w/v); χ-US-EPA Method 3050B; B_{HW}-Hot 0.01 M $CaCl_2$ soluble B

3.2. Changes in pH and EC of soil

Fly ash types and rates had a significant ($P<0.05$) effect on soil pH. Alkaline FA-II ash at 108 Mg ha^{-1} increased the soil pH by nearly 2 pH units in the top 0-10 cm of the soil. This pH increase was mainly due to the higher initial ash pH and relatively high neutralizing value of this ash. However other ash treatments had a small but consistent pH increase in the 0-10, 10-20 and 20-40 cm layers with fly ash application (Fig 1). It is interesting to note however, that the acidic fly ashes, which had virtually no liming value based on the standard calcium carbonate equivalent (CCE) test, also consistently increased the soil pH by 0.2 to 0.3 pH units. Studies by Ishak et al.[16] have also shown that with the acidic ash application rate of 10 and 50 g ash kg^{-1} to an agricultural soil, soil pH increased by 0.32 and 0.15 units respectively over the unamended control. This pH increase could possibly be due to the consumption of protons during the dissolution of glassy amorphous silicate minerals present in the fly ash as suggested by Seoane and Leiros.[17] This long-term gradual increase in soil pH may affect solubility and the bioavailability of trace element such as B, Mo and Se in soils and need to be further investigated.

All fly ashes increased soil salinity mainly in the 0-10 cm depth, with electrical conductivity (EC) rising to between 0.207 dSm^{-1} (±89) to 0.386 (±100) dSm^{-1} from a background of 0.081 dSm^{-1} (±4) in the untreated control, which indicated salt movement was slow below the root zone in these soils. This was consistent with the studies by Kukier et al.[18] in which fly ash raised the soil EC in the zone of incorporation beneath which changes in EC were minimal.

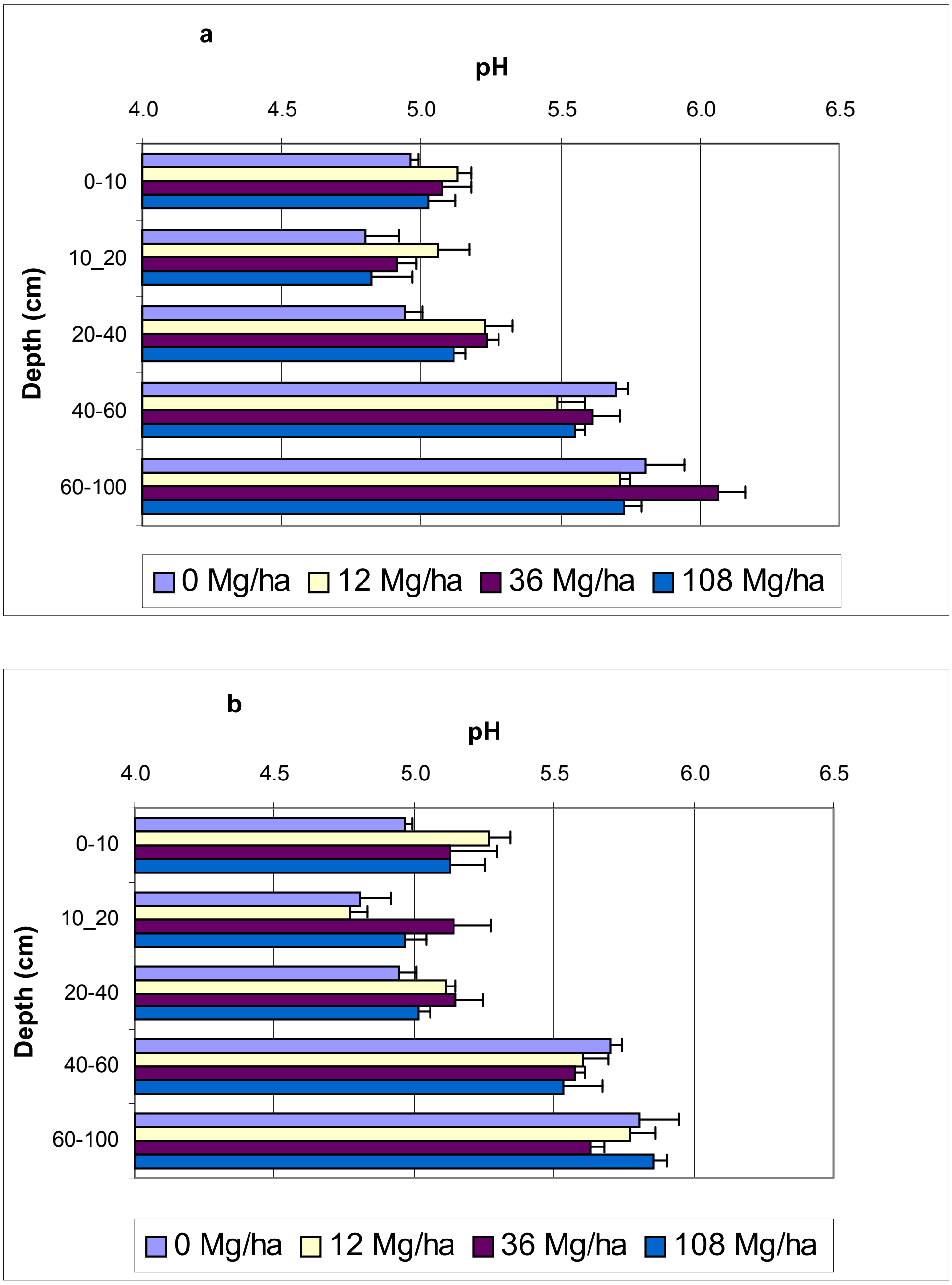
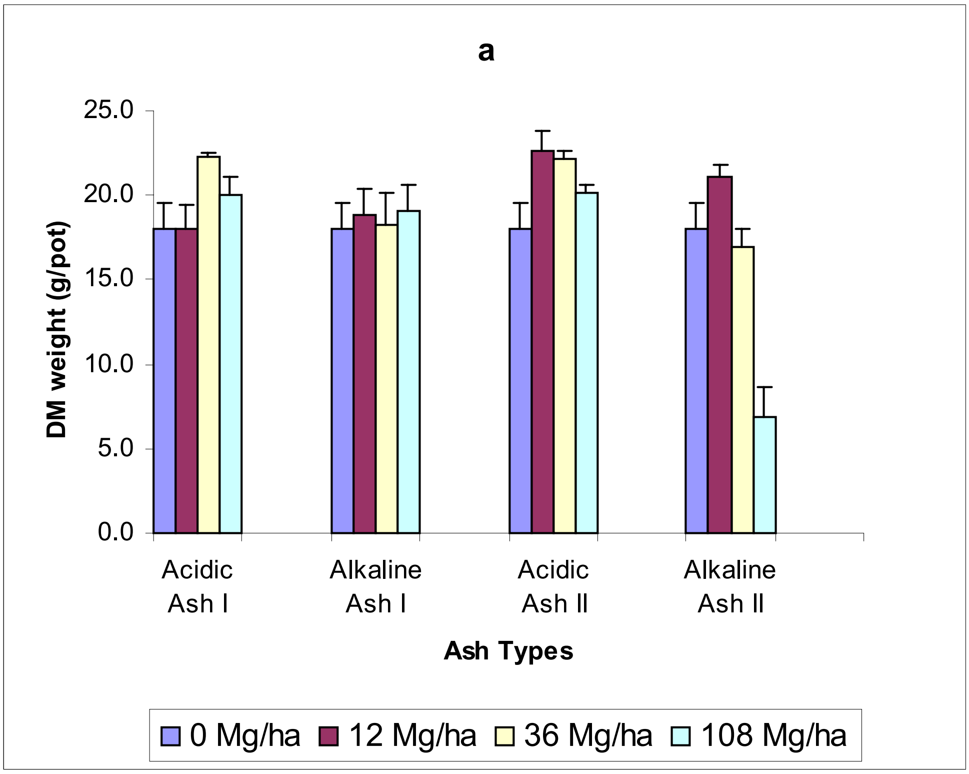


Fig. 1. Mean soil pH $_{(H2O)}$ for the acidic FA-I (a) and alkaline FA-II (b) treatments as a function of depth. (Error bars indicate 1SE)

3.3. Effects of fly ash application on crop growth and seed yield

Differences in above ground biomass (stem + pods) at the end of the 5 months growing period showed highly variable response to ash treatments (Fig. 2). This could possibly be due to the excessive variability caused by severe powdery mildews attack on the canola leaves from soon after flowering and progressively worsened until harvest.



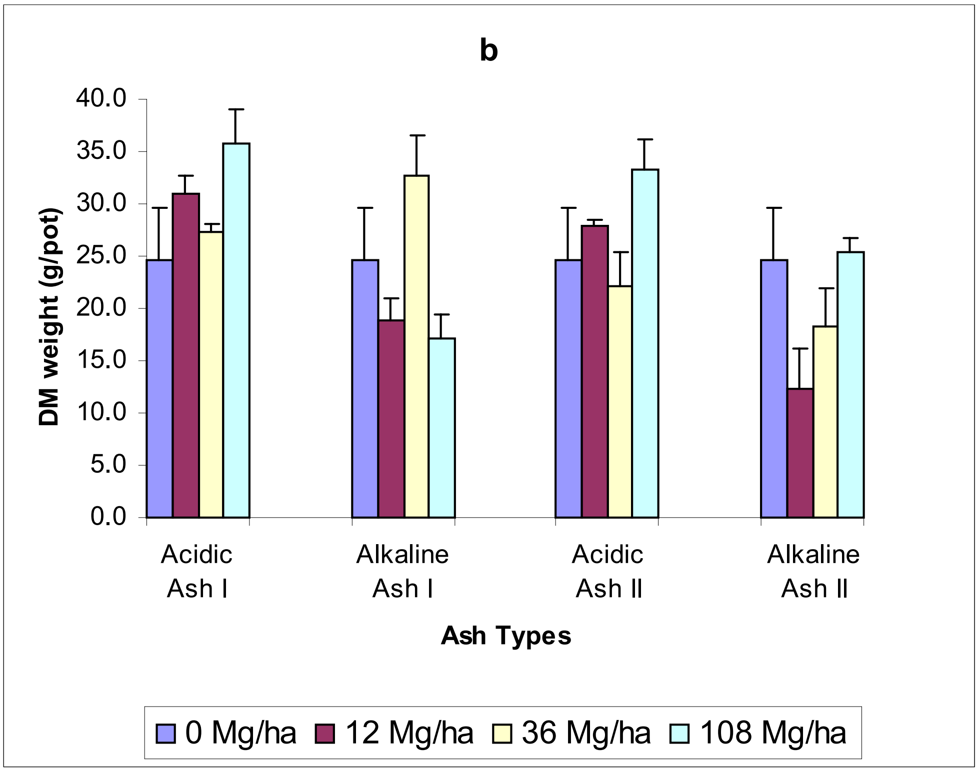


Fig.2. Mean above ground biomass of canola in response to fly ash amendment in (a) Belangalo soil (b) Kangaloon Soil (vertical bar=SE)

At the harvest only a few leaves were left on the plants due to premature defoliation associated with this disease. However, on both soils, the two acidic ashes generally increased dry matter (DM) production compared with control. Highest growth increase was observed with application of Acidic FA-I ash to Kangaloon soil and this ash had the highest extractable P. Therefore increase in DM could be possibly due to the improvement in the P nutrition of Canola plants in this mildly acidic and P deficient soil. Studies by Pathan et al.[19, 20] have also shown that fly ash application increased soil extractable P and increased plant P uptake and subsequently growth.

However, previous glasshouse studies have shown reductions in bean and corn biomass yield with increasing rates of ash addition that was primarily attributed to B toxicity.[3, 21] In the current study in which we used relatively low to high rates of ash suggesting that either canola was able to withstand high tissue B concentrations [22] and /or most of the fly ash- derived B was adsorbed by soils with high clay content and become less plant available.[23] This was supported by substantial increase in soil B in the top 0-10 cm of the soil core.

None of the fly ashes had significant effect on seed yield on Belangalo soils compared with control (Table 3). A similar response to fly ash was observed on Kangaloon soil except with Acidic FA-I ash in which seed yield increased at all rates. This yield increase could possibly due to increased soil P availability because Acidic FA-I ash had the highest extractable P (Data not shown) among all ashes used in this study and would have improved P nutrition of canola. Yunusa et al.[2] found that of 5 Mg ha^{-1} fly ash increased the canola yield by up to 25 % compared with soil-only control and yield increase was suggested be associated with increase in P uptake by canola.

Table 3. Mean seed weight (g) per plant of canola in response to fly ash application

Ash Type	Belangalo soil				Kangaloon soil			
	Ash Rates (Mg ha^{-1})							
	0	12	36	108	0	12	36	108
	1.77 (0.09)				2.13 (1.36)			
Alkaline FA-I		1.38 (0.24)	1.51 (0.32)	1.52 (0.10)		1.54 (0.54)	2.72 (0.38)	1.32 (0.23)
Acidic FA-I		1.09 (0.18)	1.87 (0.19)	1.63 (0.19)		2.55 (0.48)	2.37 (0.29)	3.15 (1.24)
Acidic FA-II		1.70 (0.20)	1.93 (0.56)	2.17 (0.8)		2.52 (0.18)	1.55 (0.66)	3.06 (0.08)
Alkaline FA-II		0.89 (0.18)	0.77 (0.22)	0.68* -		1.30 (0.77)	1.36 (0.59)	1.48 (0.82)

Values within parenthesis are standard error, n=3)- data not available* only one replicate

3.4. Effects of fly ash application on tissue B, Mo and Se concentration

Stem B concentrations were not significantly affected by ash treatments (Table 4). The exception was Alkaline FA-I ash that increased the B concentration in the stem nearly 2 fold in both soils with 108 Mg ha^{-1} compared with control but the values are within sufficiency range of 20-100 mg kg^{-1} and well below the toxic threshold limit of 200 mg kg^{-1}.[24]

Table 4. Boron, Mo and Se concentrations in the Canola stems

Soil	Ash type	0	12	36	108
			Mg ha^{-1}		
		B			
Belangalo					
	Control	7.20 (1.84)			
	FA-II		7.03 (1.22)	8.21 (0.95)	9.33 (1.91)
	Alkaline FA-I		11.17 (4.04)	8.61 (2.48)	13.26 (3.55)
	Acidic FA-II		5.68 (2.48)	3.93 (0.36)	9.27 (1.79)
	Alkaline FA-II		6.43 (0.50)	7.83 (1.59)	7.93 (1.55)
		Mo			
Belangalo	Control	0.28 (0.02)			
	Acidic FA-I		0.79 (0.20)	0.60 (0.02)	0.9 (0.09)
	Alkaline FA-I		0.52 (0.17)	0.52 (0.12)	0.86 (0.06)
	Acidic FA-II		0.35 (0.04)	0.43 (0.04)	0.69 (0.08)
	Alkaline FA-II		0.38 (0.04)	0.79 (0.26)	0.51 (0.08)
Kangaloon	Control	0.34 (0.08)			
	Acidic FA-I		0.29 (0.05)	0.54 (0.10)	0.57 (0.11)
	Alkaline FA-I		0.30 (0.04)	0.84 (0.46)	1.28 (0.43)
	Acidic FA-II		0.38 (0.10)	0.42 (0.10)	0.42 (0.03)
	Alkaline FA-II		0.48 (0.16)	0.47 (0.05)	0.70 (0.12)
		Se			
Belangalo	Control	0.08 (0.02)			
	Acidic FA-I		0.12 (0.02)	0.16 (0.02)	0.24 (0.01)
	Alkaline FA-I		0.11 (0.02)	0.10 (0.01)	0.15 (0.01)
	Acidic FA-II		0.08 (0.01)	0.09 (0.01)	0.14 (0.01)
	Alkaline FA-II		0.11 (0.01)	0.26 (0.02)	0.28 (0.05)
Kangaloon	Control	0.34 (0.02)			
	Acidic FA-I		0.46 (0.07)	0.34 (0.09)	0.37 (0.06)
	Alkaline FA-I		0.34 (0.04)	0.52 (0.07)	0.60 (0.09)
	Acidic FA-II		0.40 (0.04)	0.59 (0.11)	0.53 (0.04)
	Alkaline FA-II		0.81 (0.16)	0.59 (0.04)	0.53 (0.06)

Values within parenthesis are standard error, n=3)

It is noteworthy to mention that toxic threshold values are general guidelines only because sensitive crop may exhibit toxic responses at B levels well below the 200 mg kg^{-1}. In addition uneven distribution of B can also occur within plant parts [24] which may also hamper the direct comparison of stem B values from our study with the reported toxic limit values. However there were no visual B toxicity symptoms in canola plants growing on ash-treated soils suggesting that tissue B did not reach toxic limit to the plants in this study. However, greenhouse studies, carried out non-leaching conditions showed that excessive rates of fly ash increased B in plant leaves to phytotoxic levels.[3,25]

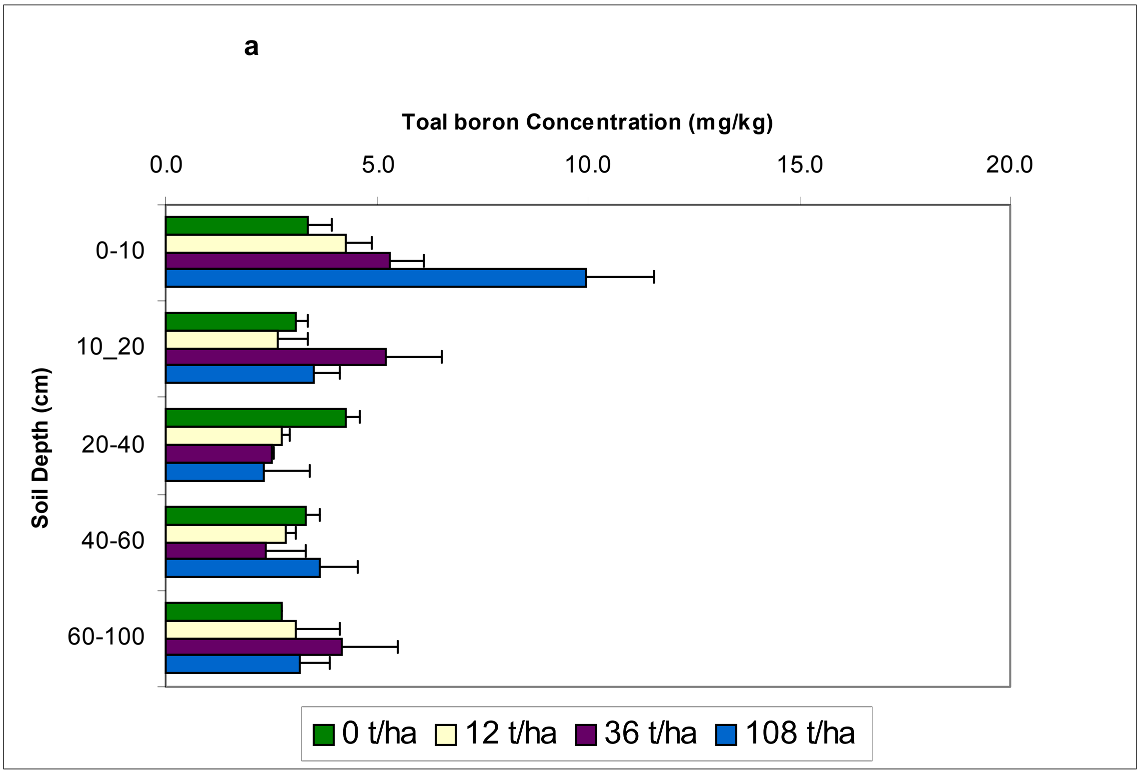
Alkaline FA-I ash applied at 108 $Mgha^{-1}$ increased the stem Mo concentration by 3 fold on both soils (Table 4). In contrast, Acidic FA-I ash, which had the highest Mo content, increased the Mo concentration in shoot in only in Belangalo soil only. This could possibly be due either Mo in the Acidic FA-I ash was less soluble than Mo in the Alkaline FA-I ash and /or increased adsorption of Mo in the Kangaloon soil due to its higher clay content. But even with the highest rate of ash additions concentration of Mo increased only up to only 1.28 mg kg^{-1} in the plant tissue, which is below the level considered to be toxic to plants or animals.[26]

Stem selenium concentration was not significantly affected by ash application. Exception was Alkaline FA-II ash at 12 Mg ha^{-1} increased the Se concentration by two fold compared with control. However, further increase in Alkaline FA-II ash addition only slightly increased the Se concentrations in the stem. This could be due to the substantial increase in soil pH (Fig 1) only at 12 Mg ha^{-1} ash addition, which might have increased Se solubility in soil and increase plant availability.[27]

High rates of alkaline fly ashes increased the seed B concentration by 45 % over control in the Kangaloon soil, however, it was not statistically significant (P=0.05) due to excessive variability (data not shown). Similar trend was observed with Mo and Se concentrations in the seeds.

3.5. Concentrations of B, Mo and Se in soil

The boron-rich Alkaline FA-I and Alkaline FA-II fly Ashes increased the soil total B substantially in the surface 0-10 cm at the application rate of 108 Mg ha^{-1} from the back ground level of 3.37 mg kg^{-1} to 9.9 and 18.8 mg kg^{-1} respectively (Fig.3.). Compared with acidic ashes, alkaline ashes have been reported to contain high amounts of B and to be responsible for B toxicity in number of plants species with fly ash application (Carlson and Adriano, 1993).[4] In the current study, soil B content below the ash incorporation layer was not affected by source of ash or rate of application in any of the ash treatments. This could possibly be due to (i) ash derived B was adsorbed on to the surface soil and/ or (ii) B in the subsurface layers might have been depleted by plant uptake. However, low plant B uptake (data not shown) suggests that most of the applied B must have been adsorbed onto the surface soil. Boron adsorption on soils mainly dependent on solution pH and B adsorption by soils increases with pH in the range of 3-9 (Goldberg, 1997).[23] At the pH of these experiments, most of the applied B must have been sorbed on to the soil since the ash material was mixed thoroughly with the soil before regular watering was started, which might have reduced B leaching. With the exception of Acidic FA-II Ash which had the lowest Se content (<1 mg kg^{-1} of total Se), all other ashes at higher rates slightly increase the Se levels in the top 10 cm of the soil. Alkaline FA-II ash, which had the highest Se content increased the soil Se levels by 3 fold compared with control. Molybdenum content in the top 10 cm of the soil was doubled by the application of Alkaline FA-I and Acidic FA-I ashes, which had high total Mo content (Table 2).



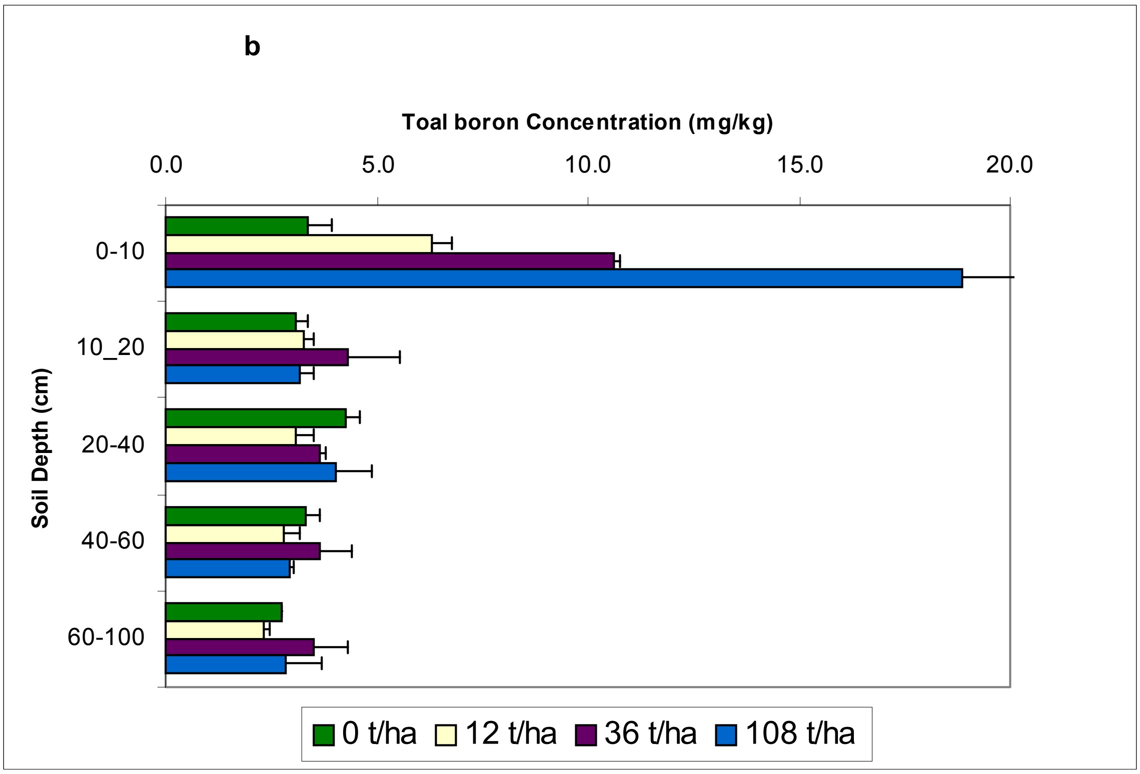


Fig.3. Total B distribution as a function of depth for the Belangalo soil applied with (a) Alkaline FA-I and (b) Alkaline FA-II (Error bar=1SE)

3.6. Effect of fly ash application on leachate B, Mo and Se Concentrations

Leachate concentrations of Mo, Se in the soil cores remained low and below detection limits throughout the leaching period. This could be possibly due to the redistribution and adsorption of these elements in the soil profile. Even the elements, which had measurable concentrations, had highly variable elemental concentration in the leachate. Since leachate elemental content is a reflection of the elemental composition of the ashes, much of the variability may be due to ash-soil interaction products, variable soil adsorption, plant uptake and water related factors.

Conclusions

The results of this study indicate that with the three of four fly-ashes tested, at rates up to 36 Mg ha^{-1}, showed no adverse effects on plant growth or accumulation of B, Mo and Se in the plant parts. Magnitude of the trace element uptake by canola varied with ash types and rates. Soil retention, redistribution and plant uptake of these trace elements might have reduced the amounts of these elements leached. Selection of crops with high B tolerance and soil types to match the ash type may allow for the successful use of high fly ash rates in agricultural soils. Although the increase in soil and leachate pH as a result of fly ash application may decrease the bioavailability of B, other elements such as Se and Mo may become more bioavailable in the long-term. These aspects of the use of fly ash need to be further investigated.

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