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CONCENTRATIONS OF SOME MAJOR AND TRACE METALS IN BOTTOM ASH OF STEM WOOD OF *ACACIA STENOPHYLLA* A. CUNN. EX BENTH. BURNT IN AN OPEN HEARTH

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ABSTRACT

The concentration of various major and trace metals (Na, K, Ca, Mg, P. Mn, Cu, Zn, B, Fe, Cd. Pb, Cr, Ni, Co, and V) were determined by means of Atomic Absorption Spectrophotometer (AAS, Perkin-Elmer Analyst 700 single beam) in the bottom ash of stem wood of *Acacia stenophylla* A. Cunn. Ex. Benth. burnt in an open hearth. The metals were in following order of concentration:

Major metals: Ca >> > K > Fe > Mg > P > Mn > Na > Zn > B > Cu

Trace metals: Pb > Cr > V > Co > Ni > Cd

The metallic contents were compared with that of some African and Australian *Acacia* species, some *Pinus* species and that in the bottom ash of some energy-generating units of European countries reported in the literature. The heavy metals contents in *A. stenophylla* bottom ash were found within permissible limits from land application viewpoint for forest nutrification in acidic soils deficient for K, Ca and P.

Key Words: Bottom ash, stem wood, *Acacia stenophylla*, Major and trace metals

INTRODUCTION

El-Juhany *et al.* (2003) tested *Acacia* species (*A. assak, A. negrii, A. seyal, A. karoo, A. ampliceps, A. stenophylla and A. salicena* cultivated in Riyadh, KSA) for charcoal production and quality of charcoal. *A. ampliceps*, *A. negrii* and *A. assak* were found to be somewhat better in charcoal quality in terms of gross heat of combustion and fixed carbon content. High calorific value of wood is not, however, the alone parameter to be viewed for fuel wood quality but elemental contents should also be taken into account as various wood types have differential metallic contents in their ash. Besides physiological and trace metals, several xenobiotic metals have also been reported from the wood ash of several species (Kumar *et al.*, 2009; Saarela *et al.*, 2005; Liodakis *et al.*, 2005; Ŝyc *et al.*, 2013; James *et al.*, 2014) which are carcinogenic and causes several other diseases.

Ash is generated as by-product of wood combustion intended for heat or power generation (Pitman, 2006). The use of biomass is common to generate energy for various uses which produces a significant volume of solid residue (Fly ash, solid ash and slag). According to Van Eijk *et al.* (2012), more than 525 k ton of ash is produced per year from wood in Europe – Austria (141 k ton per year), Denmark (> 32), Finland (100), Germany (137), Ireland (1.2), Netherlands (26), Norway (75) and Sweden (155 k ton per year) as a result of biomass utilization for energy generation. Fly ash is the lightest waste component from the burn (~ 200µm) and often has higher level of heavy metals such as Cd, Co, As, Cr, Pb, Zn and Mn. Ni is, however, in lower concentration in fly ash than bottom ash (Hakkila, 1989). Bottom ash has found several uses such as fertilizer and liming agent in agriculture and forestry, and compost and cement production (in Austria).

In Pakistan, biomass (inclusive wood) is regularly burnt for various purposes e.g., to cook food in rural areas, heating of dwellings in winter particularly in mountainous areas generally in open hearth and also in kilns in brick manufacturing, etc. Thy *et al.* (2008) have reported that wood ash is composed primarily of oxides of Ca and Si (c 60%). Mn occurs in major element concentration of 20,000 ppm. Sr and Ba are abundant (c 2200 ppm each). Elements in intermediate concentration (10-100) include many transition metals (V, Cr, Co, Ni, Cu, Zn), Zirconium and Rubidium. Elements in 1-10 ppm range include Beryllium, Arsenic, Selenium, Cadmium, tin, Lanthanum, Cerium, Neodymium and lead. The lead concentration averages 7 ppm.

Acacia stenophylla A. Cunn. Ex Benth. is a potential candidate for biosaline afforestation (Shirazi *et al.*, 2006; Sahito *et al.*, 2013). In present study, we have undertaken to investigate major and trace metallic contents in bottom ash of wood of this species when burnt in an open hearth. This is pertinent in view of the fact that several physiological and xenobiotic elements are released as fly ash or residual bottom ash from the burnt wood.

MATERIALS AND METHODS

Dry wood of stem (admeasuring 5-10 cm in diameter) was obtained from a tree of *Acacia stenophylla* growing in the Biosaline Research Field of Department of Botany, University of Karachi. It was dried in sun. The 12 kg of dry wood was burnt in air in open hearth and gray-white ash (200g in mass) was obtained.

Thermal decomposition of the biomass leads to significant weight loss in ash above 525 °C. According to Van Eijk *et al* (2014) trace metals abundances are affected by two fundamental processes 1) Enrichment resulting from volatile releases of organic and inorganic components. This enrichment can amount to as high as a 60%. It leads to increase in concentration and 2) Depletion resulting from trace element removal along with the volatile components. Depletion can also be severe that elements can be completely removed from the solid fraction. The two processes of enrichment and depletion are influenced by firing temperature. Depletion is noticeable above 800°C for alkali metals (Na, K, Rb, and Cs) and could also be seen for other elements (Cl. Ag, Cd, As, Se, and Pb). In view of this, in order to remove residual carbon, ash obtained from the hearth was re-heated in electric furnace (at 550 °C) for two hours. One gram of so treated wood ash (three replicates) was weighed on electric balance and mixed with 5 mL of conc. HNO_3 followed by addition of 10 mL de-ionized water. The solution was kept in microwave oven at 200 °C for few minutes (until the solution was clear and whitish precipitates settled down in the bottom. The solution was filtered with Whatman Filter paper and filtrate was analyzed for `major or physiological (Na, K, Ca, Mg, Mn, P, Cu, Zn, Fe, B) and trace (Cr, Ni, Pb, Cd, Co and V) elements on atomic absorption spectrophotometer (AAS) (Perkin-Elmer Analyst 700 single beam). The data was expressed as mean (mg.g^{-1} wood ash).

Table 1. Mean concentration [mg.g^{-1} dry ash ± SE, range and CV (%)] of major and trace metals in bottom ash of *A. stenophylla*.

MAJOR METALS	
Na *	0.935 ± 0.026 (0.889 - 0.979; 4.82)
K	16.292 ± 0.324 (15.675 - 16.77; 3.44)
Ca	85.779 ± 1.992 (82.784 - 89.552; 4.02)
Mg	8.664 ± 0.461 (7.779 - 9.331; 9.22)
P	1.783 ± 0.257 (1.332 - 2.221; 24.93)
Mn	1.647 ± 0.191 (1.0336 - 2.012; 20.09)
Cu	0.1397 ± 0.021 (0.099 - 0.166; 26.04)
Zn	0.351 ± 0.0198 (0.322 - 0.389; 9.77)
B	0.183 ± 0.0573 (0.13 - 0.27 (54.23)
Fe	9.506 ± 0.382 (8.752 – 9.991; 7.07)
TRACE METALS	
Cd	0.00603 ± 0.00035 (0.0055- 0.0067; 10.05)
Pb	0.0783 ± 0.0128 (0.055 – 0.099; 28.31)
Cr	0.052 ± 0.00586 (0.043 – 0.063; 19.52)
Ni	0.00703 ± 0.00549 (0.0011 – 0.018; 135.26)
Co	0.00827 ± 0.00065 (0.0073 – 0.0095; 13.61)
V	0.020 ± 0.0053 (0.010 – 0.028; 45.90)

*, Some halophytes do need Na for proper growth.

RESULTS AND DISCUSSION

The ash content of the stem wood of *A. stenophylla* was around 1.6% of the wood. Typically, the wood ash is reported to vary between 0.43 to 1.82% of the dry wood burnt (Misra *et al*., 1993). Ash contents have been reported to vary from species to species e.g., *Acacia nilotica* (2.8%), *A. leucocephala* (2.7), *Prosopis cineraria* (2.5%), *Tectona grandis* (2.2%), *Butea monosperma* (1.8%), *Sterculia urens* (1.4%) and *Cassia fistula* (1.6%) as reported by Kumar *et al.* (2009). *A. stenophylla*, in this respect, appeared to resemble *Cassia fistula*. It appears to be certain that the ash content of wood should be dependent upon the local environmental conditions of plants and their eco-physiological characteristics. Higher temperature of furnace (> 550°C) is also known to reduce the mass of the wood ash (Misra *et al*., 1993; Thy *et al*., 2008). Ash contents are also reported to decrease from upper to lower end of 3-9 years old Acacia *mangium* plants in Mozambique (Lhate, 2011). Herbs have lower heavy metal contents due to shorter rotation period, higher pH, low deposition values of metals deposition on agricultural soils, and lower metal uptake (Van Eijk *et al*., 2012).

The concentrations of various major and trace metals in *A. stenophylla* as determined by AAS are presented in Table 1. The most predominant physiological element was Calcium (c 85.78 mg.g^{-1} ash i.e. 8.6% of the wood ash) followed by Potassium (16.29 mg.g^{-1} ash, 1.63%), Iron (9.51 mg.g^{-1} ash, 0.9%) and Magnesium (8.66 mg.g^{-1} ash, 0.87%). Other physiological metals viz. Sodium, Phosphorus, Manganese, Zinc, Copper and Boron were comparatively quite lower in concentration. The metals were in following order of concentration:

Major metals: Ca >> > K > Fe > Mg > P > Mn > Na > Zn > B > Cu
Trace metals: Pb > Cr > V > Co > Ni > Cd

Table 2. Composition of wood ash (typical and for various species of plants. (mg .g^{-1}).

Metals	Wood ash (Alberta)	*Acacia karoo* Ash (South Africa) Sweet Thorn	*Acacia erioloba* ash (S. Africa) Camel Thorn	*Acacia sieberiana* ash(S. Africa) Paper Bark Thorn	*Acacia mangium* ash (Middle stem part)	Wood ash used in forest nutrition * (Croatia)	Pinus banks-iana	Pinus sylvestris
Na	48.0	0.0085 ± 0.0025	0.002 ± 0.003	0.0057±0.0060	-	-	0.023	≤ 0.022
K	42.0	0.124 ± 0.140	0.0594 ± 0.014	0.109±0.0057	-	-	0.0225	0.300
Ca	243	0.241 ± 0.043	0.243 ± 0.049	0.243±0.0410	-	34.307 ± 7.725	0.387	0.600
Mg	12.0	0.0346 ± 0.011	0.0265 ± 0.005	0.0691±0.0053	-	19.378 ± 0.0052	0.0332	0.120
P	6.0	0.0131 ± 0.0057	0.0082 ± 0.003	0.0061±0.0041	-	34.042 ± 4.750	0.0122	0.030
Mn	-	0.275 ± 0.245	0.117 ± 0.054	0.315 ± 0.0145	0.0145	11.87± 0.411	0.0390	0.070
Cu	0.032	0.083 ± 0.0326	0.096 ± 0.083	0.07 ± 0.0052	0.001	0.0977 ± 0.0029		
Zn	1.7	0.199 ± 0.098	0.173 ± 0.029	0.0969±0.0440	-	-		
B	0.038	-	-	-	-	-		
Fe	-	0.498 ± 0.256	0.584 ± 0.264	0.541±0.095	0.0563	4.235 ± 0.217	0.035	≤ 0.015
Cd	-	-	-	-	-			
Pb	0.042	-	-	-	-	0.0330± 0.00097		
Cr	-	-	-	-	-	0.0033 ± 0.00007		
Ni	0.024	-	-	-	-	0.408 ± 0.0012		
Co	-	-	-	-	-	-		
V	-	-	-	-	-	-		
Al	3.8	-	-	-	0.0463	4.018 ± 0.150	0.0333	≤0.018
Ba	1.1	-	-	-				-
S							0.104	-
Si							0.0748	-
Source	Alberta Environ. (2000)	NDLovu (2007)			Wistara and Yustiana (2014)	Marozsán *et al* (2010)	Werkelin *et al.* (2005) (Burnt at 575°C)	

Metals	Bottom Ash (Czech)	*Acacia nigrescens*† ash (Mozambique)	*Acacia aneura* (outer wood) (Australia)	*Acacia* (soft wood) (Australia)	*Pinus halepensis* ash (Greece)	*Olea europaea* ash (Greece)	Pallet Burner ash (Canada)	Wood Pallet ash (Finland)
Na	-		-	0.20	17.80	5.2	2.851	5.0
K	-		0.50	0.40	130	130.1	95.23	70.0
Ca	-		2.50	0.40	192	328	246.2	100.0
Mg	-		-	0.20	88.4	42.2	50.91	164.0
P	-		0.083	0.16	31.3	15.6	8.085	0.08
Mn	-	0.0013 ± 0.0003	-		11.2	11.5	25.845	1.370
Cu	0.995	0.0017 ± 0.003	-		-	-	0.1325	0.023
Zn	0.407	Concentration of Sb, S, Al, Fe, Na, As, Pb, P, Ba, Cd, Co, Cu, Cr, Hg, Mo, Ni, V, Sn, Zn, Se, and Ti were smaller than Cu and Mn.	-		2.8	5.0	-	0.350
B	-		-		-	-	-	0.64
Fe	-		-		9.1	16.7	6.659	-
Cd	0.00157		-		-	-	0.0077	0.016
Pb	0.115		-		-	-	0.0004	0.012
Cr	0.228		-		-	-	0.0541	0.18
Ni	0.118		-		-	-	0.0545	0.110
Co	-		-		-	-	0.0072	-
V	-		-		-	-	0.0106	-
Al	-		-		-	-	7.689	-
Ba	-		-		-	-	-	-
Source	Ŝyc et al. (2013)	Lhate (2011)	Beadle & White (1968) **	Feller (1980)	Liodakis *et al.* (2005)		James *et al.* 2014)	Kuokkanen *et al.* (2009)

*, For nutrition of forest composed of *Robinia pseudoacacia*, *Populus* x *eurqmericana* (Poplar) and *Quercus rober*. **, seen in Lambert (1981). †, Its habitat is primarily wooded grassland - particularly alluvial soil by rivers and lakes.

Table 3. Comparison of heavy metals detected in bottom ash of *A. stenophylla* with the permissible limits (mg.g^{-1} ash) of various metals in wood ash from land application viewpoint.

Metals	EPA LIMITS	Allowable Limits	Environ. Limits †	Limits as Fertilizer	Finland limits	Finnish Forest Fertilizers Limits	German Max. Limits	*A. stenophylla*
Cu	-	0.600	2.2	0.400	0.60	0.700		0.1347
Zn	-	1.50	-	7.0	1.5	4.50		0.351
B	-	-	-	-	-			0.183
Fe	-	-	-	-	-			9.510
Cd	0.001	0.0025	0.020	0.03	0.003	0.175	0.0015	0.00603
Pb	0.005	-	0.5	0.30	0.150	0.150	0.150	0.0783
Cr	0.005	0.300	1.06	0.10	-	0.30	0.002 (Cr IV)	0.0570
Ni	-	0.100	0.18	0.070	0.10	0.150	0.080	0. 00703
Co	-	-	0.150	-	-	-		0.00327
V	-	-	-	-	-	-		0.0200
Al	-	-	-		-			-
Ba	0.100	-	-	-	-	-		-
As	0.005	0.025	0.075	0.03	0.05	0.030	0.040	-
Hg	0.0002	0.001	-	0.003	0.002	-	0.001	-
Se	0.001	-	-	-	-			-
Mo	-	-	0.020	-	-	-		-
Ag	0.005	-	-	-	-			-
Source	Thy *et al.* (2008)	Kousa *et al.* (2013)	B. C. * Canada Ministry of Environ.(2008)	Anonymous (2002)	Vesterinen (2003)	MMM (2007)	Van Eijk *et al.* (2012)	Present Study

†, Environmental limits for ash intended for land application in B.C. Canada (BC Ministry of Environment 2008, *, Seen in James *et al.* (2014).

Wood ash composition is known to vary considerably due to the nature of the species, the environmental conditions of the species and the type of combustion (Alberta Environ., 2002). The variation in concentration of metals in wood ash of *A. stenophylla* was low in case of Na, K, Ca (3.11-4.82%), moderate in Mg, Zn, Fe, Co and Cd (7.07 to 13.61%) and high in case of P, Mn, Cu and Pb (20.9 to 28.31%). The variability was of very high order in case of V and B (45.9 and 54.23%, respectively) and exceptionally high in case of Ni (135.26%). It indicated to the metals heterogeneity in the wood ash.

As regards to the monovalent metals, Na and K, the wood of *A*, *stenophylla* appeared to contain K in substantially higher proportion than Na (K / Na ratio: 17.46 ± 0.698) which indicated to the potassiophillic nature of the plant as has also been indicated in previous studies (Shirazi *et al.*, 2006, Sahito *et al.*, 2013) of *A. stenophylla* saplings / seedlings grown under non-saline (control) as well as saline conditions. Another species of genus *Acacia* (A. *coriacea* subsp. *pendens*) is also reported to be potassiophillic in nature (Sahito and Khan, 2013).

Elements such as Cu, Mo, Ni, Mn, Cl, and Zn are essential for plant growth in low concentrations (Reeves and Baker, 2000; WSDA, 2007). Nevertheless, they beyond certain concentrations become toxic for most of the species (Monni *et al.*, 2000).

Vanadium, an ultra trace metal, is distributed extensively in nature and is present in almost all living organisms (Vankataraman and Sudha (2005). It was 0.020 ± 0.0053 mg.g^{-1} in ash of *A. stenophylla* i.e. in substantially higher concentration than that of other trace metals. On an average plant has vanadium in 1ppm concentration. Roots have more Vanadium than seeds and leaves (Vankataraman and Sudha, 2005; Vwioko *et al.*, 2006), The root nodules have vanadium 4 ppm and occasionally up to 12 ppm (Gummov, 2011).

Table 2 presents the metals composition of wood ash described for various acacias or bottom ash from some heat-generating units or wood ash utilized for forest nutrification. There was a great degree of variation of metals concentrations in wood ash with respect to species or that recovered from various heat generating units (using different wood lots). The metals such as Na, K, Ca, Mg and P in *A. stenophylla* were lower than that reported for Alberta wood ash (Alberta Environs., 2000), *Pinus halepensis* and *Olea europaea* from Greece (Liodakis *et al.*, 2005), pallet burner ash of Canada (James *et al.*, 2009) and that of Finland except P (Kuokkanen *et al.*, 2009). Ca and Fe in the wood ash of *A. stenophylla* were, respectively 2 and 2.35 times than that in ash used for forest nutrification in Croatia. Mg, P and Mn were comparatively lower in concentration (Marozsán *et al.,* 2010).The major metals were higher in *A. stenophylla* as compared to that in ashes of African acacias (*A. karoo, A. erioloba, A. sieberiana*) obtained from homestead fire places (NDLovu, 2007) or Australian species (*A. aneura*, and softwood of *Acacia* sp. (Beadle and White, 1968; Feller, 1980).

Cadmium and Lead were comparatively higher in *A. stenophylla* than that of Croatian ash (Merozsán, *et al.,* 2010). In comparison to the bottom ash from Czechoslovakia, Cd was lower in *A. stenophylla* and Pb, Cr, and Ni were lower in Czechoslovakian ash (Ŝyc *et al.,* 2013). Compared to the trace metals concentrations in Pallet Burner ash from Canada (James *et al.,* 2014), the ash of *A. stenophylla* showed Pb, Co, and V to be relatively in higher concentration, Cd and Ni to be relatively in lower concentration. Cr concentration was almost comparable.

Fly ash of coal is reported to contain major elements. In all fly ash, Si, Al, Fe and other elements Ti, P, S, Mg, Cl, K, Ca, Zn and Sr as major constituents (Chand *et al.,* 2009). Metal contents also vary with the particle size of the ash: lesser particle size increases Cr, Cu, As, Cd and Mo and decreases Ni, Pb and V (James *et al.,* 2014). In polluted areas, heavy metals contents are elevated (Saarela *et al.,* 2005).

There are several utilities of wood ash. It is known to improve the pH of acidic soils as Ca does in agricultural soils. Wood ash may be used as liming material (Alberta Environ., 2002). Short term effects of 0.25 and 0.5 $kg.m^{-2}$ wood ash fertilization in a field experiment in a 20-year old Scot pines stand of a nutrient poor sandy soil of arenosol nature increased pH of the soil and concentration of K, Ca, B, Mg in the soil in North Estonia which was translated into increase of these elements in plants, total carbohydrates decrease, higher accumulation of cellulose and lignin in needles (Mandre (2006). Wood ash can be used in acid soils to improve the nutrient availability and balance, however, wood ash application should be carried out when vegetation is established to minimize nutrient losses at short-term and reduce the potential risk for water bodies (Maria Gómez-Rey and Coutinho, 2012)in the area. The elevated levels of Ca, Mg and pH in wood ash indicate that it may be potential agent for soil remediation and soil fertility improvement (Knokkanen *et al.,* 2009). The solubility of heavy metals is very low; therefore there is no risk to use the wood ash in agriculture and horticulture. It should, however, be applied in reasonable amounts to avoid risk to the environment and regular quality assessment of wood ash and the soil are necessary. The effects of wood ash are primarily governed by application rate and soil type. For most forests sites studied by Marozsán *et al.* (2010), a single wood ash application per rotation could replace all nutrients lost after whole-tree harvesting except N.

Wood ash addition had significant positive effects on the tree growth, needle nutrient status and soil nutrient status, leaching water quality and carbon balance in the area. Phosphorus was the most limiting nutrient prior to the treatment in Norway spruce stands with haplic-podsol soil. No significant effects in terms of nitrogen leaching were seen (Glimtoft (2005). Permissible limits of metals in wood ash from land application view-point appearing in various publications are outlined in Table 3. The comparison of Table 1 with Table 3 makes it explicitly clear that heavy metals concentrations in ash of *A. stenophylla* were below permissible limits and it can be used for nutrification of forest land particularly in acidic soil deficient in K, Ca and P.

Mlamboi *et al.* (2011) have reported that tannin inactivation in fruits of *Acacia nilotica* and *Dichrostachys cineraria* may be achieved to a good extent by using wood ash, to improve the nutritive value of these underutilized high-tannin feeds under smallholder conditions of Zimbabwe.

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