

Analysis of Coal Ash for Trace Elements and their Geo-environmental Implications

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Abstract This study determined the content of trace elements in coal ash collected from a coal-fired thermal power plant using local coal from Sawarak, Malaysia. This is crucial for the potential impact on the geoenvironment from its disposal and utilization; as coal ash has recently been produced locally in substantial amounts and very limited data is available. The trace elements concentrations presents in coal ashes are compared with the reported coal ash concentrations and the risk for the local wet tropical geoenvironment from the perspective of its vulnerability to these is studied for an indication of potential environmental implications on the wet tropics. The trace elements were found to be in concentrations that, if applied or inadvertently released into the environmental media, present a potential hazard and further necessary research in this regard is indicated.

Keywords Trace elements · Coal ash · Wet tropics · Geoenvironmental implications

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1 Introduction

Coal combustion generates an enormous coal residue containing previously sequestered trace elements. Coal utilization in thermal power plants for electrical power generation has increased significantly and accumulating coal ash poses disposal challenges and a threat to the geoenvironment (Adriano 1986). Apart from being a continuing disposal problem their uses in soil treatment, as conditioners or filler material for low lying waste lands, in refuse dumps reclamation and construction or geotechnical secondary raw materials (Cabrera and Woolley 1994; Eymael and Cornelissen 1996; Barnes 2001; Adriano et al. 2002; Giere et al. 2003) increases their potential geoenvironmental impact. Trace elements in coal and coal ash have been discussed extensively (Adriano 1986; Eary et al. 1990; Mattigold et al. 1990; Swaine 1990; Finkelman 1994) and their presence is an environmental concern (Hansen et al. 2002, Reijnders 2005). The trace elements in coal ash need to be determined and assessed for potential effects (Giere et al. 2003). As, Cr, Pb, V, Zn in coal ashes present a significant environmental burden (Reijnders 2005). The bioavailability and toxicity of trace elements (Depledge et al. 1998) is, however, dependent upon its speciation.

The trace elements poses a higher threat in the wet tropics as they can retard or are fatal for microbial activity in turn affecting nutrients supply to vegetation (Doelman 1984). Arthur et al. (1984) reported significant reductions in microbial respiration upon fly ash

addition in soil to trace metals present at potentially toxic concentrations. The microorganism removal also ceases detoxification of trace elements by enzymatic reductive processes. The organic constituents, even in small amounts, can assert a significant influence on aquatic systems (Hem 1985). The average dissolved organic content (DOC) in tropical rivers is higher than rivers in temperate, arid or semi-arid regions except the subarctic (6 mg/L as compared to 3 and 19 mg/L, respectively) (Meybeck 1982) presenting a higher prospect for interaction with trace elements. Natural organic matter (NOM) comprising of polyelectrolyte molecules is a significant component of freshwater environments and these organic surfaces can be effective trace element sorbents at much lower pH (Warren and Haack 2001). The major impact of coal ash leachates on receiving waters, apart from increased elemental concentrations, cause changes in water pH with implications for trace element mobility (Carlson and Adriano 1993). The disruption of the microbially controlled plant tissue degradation also has implications on the organic matter constitution of the aquatic environment highly dependent on supply from soils of a watershed.

Paddy soils form a special type of tropical aquatic ecosystem, occurring due to various types of submerged or waterlogged soils are very vulnerable to trace elements as their cyclic submerged and dry periods causing alternate reductive and oxidative conditions which have implications for heavy metal mobility. Large amounts of organic matter accumulate in the paddy soils unlike tropical soils because its decomposition is rigorously inhibited as it remains in a reduced condition. The substances released in the reduced layer are either adsorbed onto the soil surface or remain dissolved in soil solution available to plants (Kyuma 2004).

Studies on coal ash are important as coal utilization, mainly in thermal power plants, has increased significantly in Malaysia from 4.2 to 13 m tonnes from 2000 to 2005 (Mohd Annas 2005). As coal ash is a recent issue in Malaysia; there are limited studies on coal ash and the trace elements composition. Hence, this study determined the trace element concentrations in coal ash from a thermal power plant using local coal from Sarawak, Malaysia, classified by Wood (1974) as toxic and relatively accessible. The present study compares the concentrations described for coal ash in literature, normal levels in

environmental media and discusses their potential implications for a wet tropical environment especially the paddy soils and organic matter.

2 Materials and Methods

Coal ash samples in this study were collected from Sejingkat Thermal Power Plant, Kuching, Malaysia. The original coal sample from Kapit, Sarawak and a total of eight coal ash samples were analyzed for the trace elements present in it. The coal ash samples constituted of four fly ash samples, designated as FA-01, FA-02, FA-03 and FA-04, from the Electro-Static Precipitator (ESP); two bottom ash samples, designated as FA-05 and FA-06, from the bottom of the furnace; and two pond or lagoon ash samples, designated as FA-07 and FA-08, from the disposal lagoon. A laser particle size analyzer (Coulter LS100Q) has been employed to study the Particle-Size Distribution (PSD) of the coal ash samples. The coal ash samples have been evaluated for their mineralogical composition by conducting X-Ray Diffraction (XRD) spectrometer studies, using a graphite monochromator and Cu-K α radiation (scanned from 0 to 80°). Data files presented by Joint Committee on Powder Diffraction Standards (JCPDS 1994) were used to identify the minerals present in the coal ash samples. The trace elements present in the coal ash samples were determined by Inductively Coupled Plasma-Atomic Emission Spectrometry (ICP-AES).

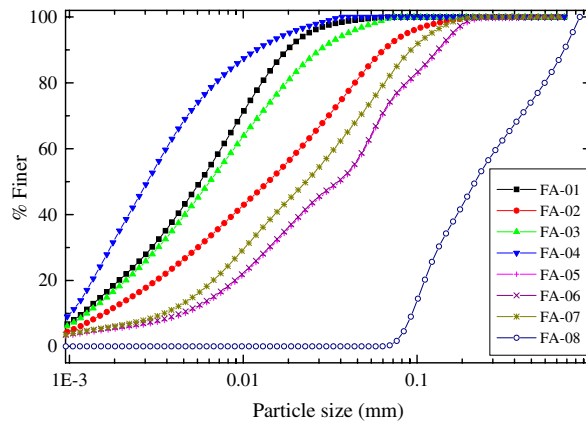
3 Results and Discussion

The results of particle size distribution (PSD) have been presented in Fig. 1. Various size fractions such as clay-sized (<0.002 mm), silt-sized (0.002 to 0.075 mm) and sand-sized (0.075 to 4.75 mm); Uniformity co-efficient (C_u) & Co-efficient of curvature (C_c) are presented in Table 1.

From the Table 1, it can be noticed that fly ash is predominantly silt-sized material with very little amount sand-sized particles. Bottom ash is silty sand sized and lagoon ash is sandy silt sized materials.

X-Ray Diffraction results shows that, quartz is the most predominant crystalline phases present in the coal ash samples and calcium oxide, iron oxide hydrate, & aluminium iron oxide are also present in all coal ash samples.

Fig. 1 Particle size distribution of the coal ash samples



The trace elements analyzed in the coal ash were As, Ba, Cr, Hg, Mo, Ni, Pb, Se, V, Zn and Cu. These were chosen because as mentioned by Reijnders (2005) and in literature that they represent a significant environmental burden. The results of trace elements concentrations from this study are presented in Table 2, which is the average of three tests results, and compared with those in other coal ashes, soils and surface waters reported in literature. Soils, plants and aquatic organisms are then considered from the perspective of their vulnerability *vis a vis* the concentration values of trace elements detected in the ash in tandem with reported impact levels to obtain an overall indication of the environmental impact.

3.1 Comparison with Trace Element Levels in Coal Ash

The reported ranges and the average values of the trace elements determined in this study for coal ash samples in literature are shown in Table 2. The comparison of the concentrations determined in this

study with the average values reported in literature showed that the concentration of As was above; while Ba was much below the average amounts. The higher values for Cr, Cu, Se and Zn (showing a wide variation enabling bunching into high and low groups) were much above the average amount. Pb was significantly lower than the average amount. Although lower a concentration with a factor of 3 higher than the average for Ni was detected. High values of Hg were detected but values were not available from literature for comparison. The solitary ‘quantitative’ reference of Hg in fly ash found only referred to ‘amounts’ was in Swaine (1990). The V detected was mostly much below the average. Only Mo was found in negligible amounts (except in one sample) although it is reported in coal ashes.

3.2 The Trace Element Levels and Environmental Implications

To determine the reference levels of trace elements, it is firstly necessary to know their contents under natural

Table 1 Particle size distribution of the coal ash samples

Sample no.	Clay-sized (%)	Silt-sized (%)	Sand-sized (%)	Uniformity co-efficient, C_u	Co-efficient of curvature, C_c
FA-01	20.80	79.20	0.00	6.25	5.63E-05
FA-02	13.53	79.66	6.81	13.44	4.06E-04
FA-03	19.15	80.29	0.56	6.62	6.77E-05
FA-04	35.84	64.16	0.00	3.60	1.04E-05
FA-05	5.51	64.89	29.60	9.27	0.88
FA-06	6.00	63.00	31.00	11.88	0.99
FA-07	6.25	71.60	22.15	10.00	0.97
FA-08	0.00	1.36	98.64	3.03	0.63

Table 2 Trace elements (in mg/kg) from the present study and reported concentrations in coal ash

Trace Element	Coal sample	Different samples used in this study								Reported in literature	
		Fly ash				Bottom ash		Pond ash		Range	Average
		FA-01	FA-02	FA-03	FA-04	FA-05	FA-06	FA-07	FA-08		
As	16.2	<1.0	<1.0	<1.0	16	14	06	<1.0	16	2.8–6,300 ^a	9 ^a
Ba	25.9	744	814	669	876	662	445	850	802	150–3,000 ^b ; 0.011–0.5% ^a	0.3% ^a
Cr	7.72	319	367	487	20	18	16	460	36	4–433 ^b ; 10–690 ^a	54 ^a
Hg	<0.5	312	<1.0	<1.0	<1.0	<1.0	<1.0	300	<1.0	13–171 ^b ; 14–1,000 ^a	63 ^a
Mo	<0.5	<1.0	1	<1.0	<1.0	<1.0	<1.0	<1.0	<1.0	7–279 ^b	48 ^a
Ni	6.8	<1.0	23	78	9.9	10	8.3	23	15	11–125 ^b ; 7–117 ^a	10 ^a
Pb	7.64	<1.0	<1.0	<1.0	6.9	5.2	2.5	<1.0	7.3	–	–
Se	14.9	603	542	516	<1.0	<1.0	<1.0	530	<1.0	14–767 ^b ; 10–4,300 ^a	34 ^a
V	<0.5	76	58	55	56	43	22	49	52	0.2–134 ^a	7.6 ^a
Zn	582	183	191	318	9.8	12	11	155	14	94–429 ^b ; 50–1,000 ^a	119 ^a
Cu	0.91	354	282	364	8.5	6.6	3.1	293	9.8	100–1,333 ^b ; 35–1,333 ^a	72 ^a

^a PECH (1980)^b Abernethy et al. (1969)

conditions (Navas and Machin 2002). The background (normal) or mean concentrations of the trace elements by various researchers for soils (unless inordinately influenced by parent materials) and natural waters are given in Table 3. The quantitative levels used in various ‘approaches’ to assess risks and effects of trace elements on plants and aquatic organisms are given in Table 4.

It is apparent that the trace elements present in the coal ash pose a prospective hazard detrimental if inappropriately disposed or accidentally released into the geoenvironment. Some pertinent implications are discussed below.

3.2.1 Plants and Paddy Soils

The Cr and Pb present on coal ash particle surface will leach into soil solution to enhance their availability to plants (Natusch and Taylor 1980; Davies 1995). A mean value of 115, 11, 2, 56, 146 and 3.3 ppm for Cr, Cu, Ni, Zn, V and Mo, respectively are reported for East Malaysian paddy soils (Domingo and Kyuma 1983). The levels shown from the coal ash in this study for Cr, Cu, Ni, Se and Zn exceed levels shown in Table 1; indicating the vulnerability of plants to phytotoxicity if accidentally or intentionally applied to soils or used as conditioners. The presence of 35–177 ppm of Cr in stem and leaf in paddy growing areas can result in a 10% yield reduction (Chino 1981). In the case of Se in a humid region selenite [SeO₃²⁻ and (Se IV)] is reported as the predominant mobile inorganic form in

soils (Cary et al. 1967) so should be expected to be found. Paddy soils retain more Hg than other soils so this is a significant concern (Kitagishi and Yamane 1981) and the high values detected in the coal ash will make more Hg available. The proposed maximum limit of As in paddy soils given by Kitagishi and Yamane (1981) is 15 ppm the coal ash exceeds phytotoxicity levels and are potentially hazardous. The level of Mo and V present in the coal ash studied does not exceed the means reported for East Malaysian paddy soils or the minimum phytotoxic level. However, accumulation from continuous loading of fly ash could lead to livestock developing fatal ‘molybdenosis’ upon ingesting forages with Mo levels at 10–20 ppm (Neathery and Miller 1977).

3.2.2 Aquatic Effects

The toxicity of aquatic organisms to trace elements has been well reviewed (Mance 1987; Wright and Welbourn 2002). Freshwater species are liable to be adversely impacted by the leaching of Cu, Pb, Ni, V and Hg from the ash. The Hg showed two high values (312 and 300 ppm). However, as concentrations of Hg at the ppb level are critical the coal ash requires to be analyzed at lower detection limits. The introduction of Ba levels found in the ash into water renders it unsuitable for human consumption. Mo was not seen to be present in high levels (as only one sample of 1 ppm was detected). Trace elements are prone to

Table 3 Concentrations of the trace elements in soils (mg/kg) and natural waters ($\mu\text{g/L}$)

Element	Soils		Fresh water
	Normal	Mean	Normal
As	0.1–40 ^a	10 ^f ; 7 ^v	0.15–0.45 ^m ; 1 ⁿ
Ba	100–3,000 ^a	82–838 ⁱ	–
Cr	10–90 ^b ; 5–1,500 ^a	84 ^g ; 50 ^f	1 ^o ; 1–10 ^a
Cu	2–60 ^w	20 ^h ; 30 ^e	c.2 ^p
Pb	10–150 ^w	17 ⁱ ; 29 ^g	0.2 ^o ; 1–10 ^g
Mo	1–5 ^w	1.5 ^f	0.03–10 ^a ; 1 ^q
Hg	100 ppb ^c ; 0.01–0.5 ^a	400 ppb ^j	–
Ni	2–100 ^w	20 ^k ; 34 ^g ; 25 ^f	1–3 ^r ; 5 ^s ; 15–20 ^t
Se	0.1–5 ^a	–	<10 ^u
V	3–500 ^a ; 20–500 ^d	100 ^l ; 90 ^j	20 ^p
Zn	25–200 ^w	50 ^c	5–15 ⁿ

^a Bowen (1979)^b Gaughhofer and Bianchi (1991)^c Adriano (1986)^d Swaine (1955)^e Lindsay (1972)^f Berrow and Reaves (1984)^g Ure and Berrow (1982)^h Baker and Chesnin (1975)ⁱ Nriagu (1978)^j Kabata-Pendias and Pendias (1984)^k McGrath (1995)^l Vinogradov (1959)^m Ferguson and Gravis (1972)ⁿ Moore and Ramamoorthy (1984)^o Trefry and Presley (1976)^p Dojlido and Best (1993)^q Kharkar et al. (1968)^r Nriagu (1980)^s Kabata-Pendias and Pendias (1979)^t Sunderman and Oskarsson (1991)^u Standard Methods (1989)^v Leonard (1991)^w Bowie and Thornton (1985)

undergo bioaccumulation in aquatic organisms. A few examples from the trace elements detected are Hg (e.g. as organo-mercury compounds), Pb (bioaccumulation factors between 1,500 and 24,000) (Salanki et al. 1982) and As (accumulates in the food chain up to 25 mg/kg) (Wright and Welbourn 2002). It is essential to determine, particularly in water, the speciation especially the free ion and free hydrated ion forms as these are the most available and toxic (Luoma 1983).

As seafood is a substantial component of the diet in this region, so priority should be to detect As⁵⁺ and Cr⁶⁺ having a higher affinity for biological binding sites and Cu(I), Cu(II) and Zn²⁺ being biologically available as As, Cr, Cu and Zn are present.

4 Conclusions

This preliminary study shows that potential implications exist from the trace elements present in the local coal

Table 4 Quantitative levels of trace elements in plants (mg/kg) and aquatic organism ($\mu\text{g/L}$) to assess risks and effects

Element	Plants	Aquatic organisms
As	20–50 ^{a,e}	50 ^{c,i}
Ba	–	550 and 600 ^{d,j} ; 1 ^{c,j}
Cr	75–100 ^{a,e} ; 100 ^{b,f}	70.05 ^c
Cu	60–125 ^{a,e} ; 20 ^{b,f}	81–5 ^b
Pb	100–400 ^{a,e}	100 and 5,000 ^{b,m} ; 100–15,000 ^{b,n}
Mo	2–10 ^{a,e} ; 6 ^{c,g}	0.04 ^{c,o}
Hg	0.3–5 ^{a,e}	12>0.5
Ni	100 ^{a,e} ; 50 ^{b,f}	0.1–9.5 ^{b,m} ; LC ₅₀ values: 50 $\mu\text{g/L}$ –11 mg/L ^{b,q}
Se	5–10 ^{a,e}	–
V	50–100 ^{a,e} ; 140 ^{b,h}	20 (in soft water) 60 (in hard water) ^{c,j}
Zn	70–400 ^{a,e}	180 (in soft water) 570 (in hard water) ^{c,p}

^a Critical soil total concentration range above which toxicity is considered possible^b Toxic level^c Permissible levels^d Fatal dose^e Kabata-Pendias and Pendias (1992)^f Adriano (1986)^g Neathery and Miller (1977)^h Gough et al. (1979)ⁱ WHO (1981)^j Dojlido and Best (1993)^k WHO-IPCS (1988)^l US EPA (1985)^m Moore and Ramamoorthy (1984)ⁿ Harrison and Laxen (1981)^o Siegel (2002)^p Wright and Welbourn (2002)^q Sunderman and Oskarsson (1991)

ash. The high concentrations of Cr, Cu, Pb, Ni, V, Zn and Se in the coal ash studied pose a hazard if they are disposed on, or applied to soils, or discharged into the freshwater. There also exists the risk of accumulation in soils from continuous loading. The importance for determining Hg is emphasized as its detection in the coal ash shows that not all of it is released to the atmosphere as commonly assumed (Swaine 1990). For Se although the bioavailable water-soluble fraction of Se poses a direct risk to plants and livestock (Kabata-Pendias and Pendias 1984) the total soil concentration, however, is sufficient to be determined as this gives a better measure of plant response than the soluble fractions (Sippola 1979).

As a consequence it is amply evident that there are areas that require further investigations. These include the need to determine speciation for bioavailability and aspects of trace elements behaviour *vis-a-vis* wet tropics i.e. detailed behavior of interaction with organics in the soils and impacts on microorganisms i.e. between microbes and organic matter bonded with trace metals subsequently effecting nutrient availability as these are two such instances where sparse information is available. From the perspective of industrial ecology (Ayers 1992) the source reduction to limit the leachable trace elements in ashes forms the optimal practice in preventing environmental hazards. The application of forced leaching is advocated as one way for the improvement of the environmental quality of the coal ash (Nugteren et al. 2001; Pederson et al. 2003) as studies have shown that the overall environmental burden that ashes pose is lowered (Reijnders 2005).

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